

*GENERATION, AMPLIFICATION, AND DETECTION OF INFRARED AND OPTICAL RADIATION
BY QUANTUM-MECHANICAL SYSTEMS*

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INTRODUCTION

ELECTROMAGNETIC radiation is produced by three kinds of sources: thermal radiation, fluorescence, and oscillators. The spectral distributions of the radiation from these sources are quite different. The spectral distribution from a thermal source is approximately the same as that of a black body, while the spectrum of a fluorescent source is much narrower than that of a black body. Oscillators, on the other hand, produce essentially monochromatic radiation.

The radiation intensity of an absolutely black body and its spectral distribution are described completely by the Planck formula, where the single parameter upon which both intensity and spectral distribution depend is the temperature. The intensity and spectral distribution of a fluorescent source cannot, however, be characterized by a single parameter (for instance, temperature). It is possible, on the other hand, to describe fluorescence radiation in terms of an effective temperature: the effective temperature of a fluorescent source is defined as the temperature of a black body which produces the same intensity within some frequency interval. This definition is not unique since it depends on the size of the frequency interval. It is also obvious that when defined this way the effective temperature depends on frequency, i.e., the effective temperature is different in different portions of the spectrum.

Because the spectral distribution of a fluorescent

source is considerably narrower than that of a thermal source, the effective temperature of a fluorescent source is considerably higher. In general the effective temperature associated with the radiation from a fluorescent material is much higher than the actual temperature of the material — that is to say, fluorescent materials radiate "cold light."

Since they produce essentially monochromatic radiation, oscillators are usually characterized by the frequency and power of the radiation. Oscillators can also be characterized by an effective temperature, just as fluorescent sources. Because of the extremely narrow bandwidth and high power of the radiation they produce, the effective temperatures of oscillators are characteristically very high. For example, oscillators in the centimeter range produce radiation with an effective temperature of $10^{15} - 10^{25}$ degrees.

Oscillators are conventionally used in the long-wave region (radio waves, centimeter waves); on the other hand, in the submillimeter, infrared, and optical regions, the principal radiation sources are thermal sources and fluorescent sources. At the present time, no one has been successful in building an oscillator in the infrared or optical regions, by electronic techniques or any other methods.

In recent years, methods of generating and amplifying radio waves by induced transitions in quantum-mechanical systems (molecules, atoms, ions, etc.)^{1-4,9} have been developed to a high degree.

The development of molecular oscillators and para-

magnetic amplifiers (masers) was stimulated by the appearance of papers that proposed the use of induced transitions for the generation and amplification of sub-millimeter, infrared, and optical waves. An analysis of these methods is given in the present review.

The radiation mechanism in masers is fundamentally different from the mechanism in fluorescent and thermal sources. Quanta are emitted by hot bodies and fluorescent sources as a result of spontaneous transitions of the atoms, molecules, etc. from high (excited) levels to levels of lower energy. Quantum emission occurs in the absence of quanta of the same frequency, being caused by the zero-point vibrations of the electromagnetic field. Because the probability of spontaneous emission falls off at lower frequencies, the quantum fluorescence yield in the infrared region is quite small. This is probably the reason why useful fluorescent materials for the infrared have not been successfully developed at the present time.

Thus, to obtain radiation in the infrared (especially in the far infrared) it is necessary to use induced transitions; the probability of quantum emission is considerably higher in induced transitions.

In an oscillator or amplifier in which induced transitions are used, it is necessary to produce states which are not thermodynamic equilibrium states; these are called negative-temperature states. In systems in thermodynamic equilibrium, and systems characterized by positive temperatures, the populations of the energy levels are smaller in higher energy levels. On the other hand, in negative temperature states, (even if over a limited energy range) there is an inverse distribution of populations over the levels: the populations of the energy levels increase with energy. In negative-temperature states quanta can be emitted under the effect of radiation incident from external sources. Thus, quantum-mechanical systems in negative-temperature states can be used as radiation amplifiers or oscillators. Although the sensitivity of quantum-mechanical amplifiers is reduced at higher frequencies because of spontaneous emission, it is reasonable to expect that a considerable increase in sensitivity can be achieved at infrared wavelengths as compared with that of existing devices used in this wavelength range. In references 5 and 6 it has been proposed to use spectral lines of various materials in high-sensitivity radiation detectors which would be free from noise due to spontaneous emission.

One of the most important parameters in a negative-temperature system is the "number of active particles," i.e., the difference in the number of systems produced per unit time in the upper level and the lower level. This number determines the power which can be radiated by the system.

Another important parameter is the quantity

$\kappa = \frac{2\pi |d|^2 n}{\hbar \Delta \omega}$ where $|d|^2$ is the square of the dipole-moment matrix element between the levels considered,

n is the number of active systems and $\Delta \omega$ is the width of the spectral line. As will be shown in Sec. 8, κ enters into the oscillation condition for oscillators and also determines the gain of amplifiers. Because molecules in gases are distributed over a large number of rotational levels, in the centimeter range κ is a thousand times smaller for gases than for paramagnetic ions in crystals, in spite of the fact that dipole moments of paramagnetic ions are a hundred times smaller. These relations obviously also hold for the infrared. For this reason solids are the most promising working materials for the generation and amplification of infrared waves.

The development of coherent sources of infrared and optical radiation is a pressing problem. For instance, the availability of coherent sources in the optical and infrared regions would make possible a tremendous increase in the sensitivity and resolving power of spectroscopic devices. Infrared and optical waves can also be used, like radio waves, for the transmission of information (the volume of transmitted information increases as the frequency of the radiation is increased and decreases as the spectral width is reduced) etc.

In the present paper we shall review all papers published in recent years in which suggestions have been made for obtaining negative temperatures in gases and solids; we shall also discuss the features of oscillators and amplifiers that operate at infrared and optical wavelengths. The last section of the second part of the paper is devoted to quantum-mechanical radiation detectors.

I. METHODS OF OBTAINING NEGATIVE-TEMPERATURE STATES

1. Negative Temperature

At the present time the concept of a negative temperature is widely used in the description of maser oscillators and amplifiers, because systems in negative-temperature states amplify, by stimulated emission, electromagnetic radiation which is incident upon them.

The conditions under which "negative absorption" of radiation occurs were first formulated by V. A. Fabrikant.⁷

The concept of a negative temperature was first introduced^{7a} to describe a nonequilibrium distribution of Li nuclei over Zeeman levels when the number of nuclei in a higher energy level is greater than the number in a lower level.

The concept of a negative temperature is widely used at present to describe the operation of maser oscillators and amplifiers.

Suppose that a quantum-mechanical system has two energy levels and that each level contains an arbitrary population (Fig. 1). At thermodynamic equilibrium the distribution of population over the levels is given by

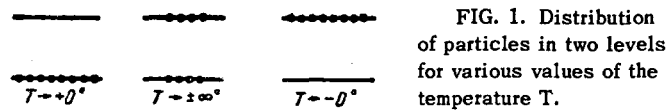


FIG. 1. Distribution of particles in two levels for various values of the temperature T .

the Boltzmann distribution function

$$N_i = A e^{-\frac{\epsilon_i}{kT}}, \quad (1.1)$$

where A is a normalization factor, N_i is the population of the i -th level, ϵ_i is the energy of the i -th level, k is the Boltzmann constant and T is the absolute temperature. It follows from Eq. (1.1) that the population of the higher energy level is smaller than that of the lower level for any value of the temperature.

In the absence of thermodynamic equilibrium the population distribution between the two levels can be arbitrary. When the population in the higher energy level is smaller than in the lower level the system can be described formally by a function of the form in Eq. (1.1) with some positive temperature T . To describe a nonequilibrium system, in which there is a larger population in the higher energy level, T must be negative because of the form of the function in Eq. (1.1). Hence, states of the system in which the upper level has a larger population than the lower level are called negative temperature states.

When the entire population is in the upper energy level, the system can be characterized by a temperature $T \rightarrow -0^\circ \text{K}$ and when the entire population is in the lower energy level the state of the system corresponds to a temperature $T \rightarrow +0^\circ \text{K}$. When the upper and lower levels are equally populated the state of the system corresponds to a temperature $T \rightarrow \pm \infty$. It should be noted that the system energy is a minimum at $T \rightarrow +0^\circ \text{K}$ and a maximum at $T \rightarrow -0^\circ \text{K}$.

The negative temperature concept can be extended to systems with an arbitrary number of levels; in these cases the distribution of population over the quantum-mechanical states of the system is described by a distribution function f_r . The distribution function f_r is proportional to the probability of finding elements of the system in the r -th quantum-mechanical state, and is normalized as follows:

$$\sum_r f_r = N, \quad (1.1')$$

where N is the total population of the system and the summation is taken over all states of the system. All systems that can be described by distribution functions fall into two classes:

1. Systems in which the probability of finding elements in a quantum-mechanical state characterized by an energy ϵ is reduced as the energy is increased over the entire range of states, i.e.,

$$f_l > f_m, \text{ if } \epsilon_l < \epsilon_m, \quad (1.2)$$

where the subscripts l and m take on all possible values.

2. Systems in which the following inequality holds for the levels ϵ_l and ϵ_m

$$f_l < f_m, \text{ if } \epsilon_l < \epsilon_m. \quad (1.3)$$

In systems of the first class, in particular, systems in thermodynamic equilibrium, the distribution function $f(\epsilon_r, T)$ is either the Fermi-Dirac distribution function or the Bose-Einstein distribution function, depending on the statistics which characterizes the elements of the system:

$$f(\epsilon_r, T) = g_r (e^{\frac{\epsilon_r - \mu}{kT}} \pm 1)^{-1}, \quad (1.4)$$

where g_r is the degeneracy of a given energy level and μ is the chemical potential.

In thermodynamic equilibrium the only parameter in Eq. (1.4) is the absolute temperature T , which is always positive.

In nonequilibrium states of a system the distribution of population over levels cannot be characterized by a single parameter (temperature). However, for a system of the first class, the population distribution for any two levels can be characterized by a function such as (1.4) with a positive value for T . These states of the system are called positive-temperature states.

In systems of the second class, which are always nonequilibrium systems, the population distribution for the levels corresponding to the inequality in (1.3) can be described formally by a function of the form given in (1.4) if the parameter T (temperature) is negative. Systems which satisfy (1.3) are called negative-temperature systems with respect to the levels ϵ_l and ϵ_m .

Systems of the first class and second class interact differently with monochromatic electromagnetic radiation at a frequency

$$\omega = \frac{\epsilon_m - \epsilon_l}{h}. \quad (1.5)$$

A positive-temperature system absorbs the electromagnetic radiation incident upon it, while a negative temperature system (with respect to the levels ϵ_m and ϵ_l) amplifies radiation if this radiation is at a frequency $\omega = (\epsilon_m - \epsilon_l)/h$.

The number of photons emitted by the system per second in making a transition from the state characterized by m to the state characterized by l is

$$I^+ = \omega_{ml} (n_\lambda + 1) f_m (1 \mp f_l). \quad (1.6)$$

The upper sign corresponds to Fermi statistics, the lower sign corresponds to Bose statistics; ω_{ml} is the quantum-mechanical probability for emission of a photon and n_λ is the number of photons associated with the radiation oscillator denoted by λ .

The number of photons absorbed by the system per second is

$$I^- = \omega_{lm} n_\lambda f_l (1 \mp f_m). \quad (1.7)$$

Subtracting (1.7) from (1.6), and taking account of the fact that $\omega_{ml} = \omega_{lm}$, we have

$$I^+ - I^- = \omega_{lm} \{n_\lambda (f_m - f_l) + f_m (1 \mp f_l)\}. \quad (1.8)$$

The first term in Eq. (1.8) is proportional to the number of photons and is to be associated with stimulated emission or resonance absorption. On the other hand, the second term (1.8), which is independent of the number of photons, characterizes the spontaneous transitions in the system. In order for the system to amplify incident electromagnetic radiation, it follows from Eq. (1.8) that the following condition must be satisfied:

$$f_m - f_l > 0, \quad (1.9)$$

that is to say, the system must be in a negative temperature state with respect to the levels m and l ($\epsilon_m > \epsilon_l$). The condition given in Eq. (1.9) is necessary but generally not sufficient for amplification because there may be other pairs of levels in the system for which the transition frequency ω is the same, but for which the temperature is positive.

Thus, the sufficient condition for amplification is

$$\sum_{m,l} \omega_{ml} (f_m - f_l) > 0, \quad (1.10)$$

where the summation is carried out over all pairs of indices for which $\epsilon_m - \epsilon_l = \hbar\omega$.

The spontaneous emission represented by the second term in Eq. (1.8) is responsible for the noise in quantum-mechanical amplifiers,^{3-5,9-11,61} since the strength of the spontaneous emission is independent of the number of photons incident upon the system.

Below we shall consider methods which can be used to produce negative temperatures in the infrared and optical regions.

2. Selection of Molecules in Molecular Beams by Means of Inhomogeneous Electric or Magnetic Fields

The method of selecting molecules by levels, used in ammonia masers,^{1,49} can also be used for selecting molecules in the infrared and optical regions.^{12,13} For example, the transition frequency between various rotational levels $j \rightarrow j+1$ of the ammonia molecule, NH_3 , is given by the formula

$$\omega = 2B(j+1), \quad (2.1)$$

where B is the rotational constant of the molecule (Fig. 2).



FIG. 2. Splitting of rotational levels into two inversion sublevels. The arrow indicates an allowed transition.

The constant $B = 2.98 \times 10^5$ Mc/sec for the NH_3 molecule. For ND_3 , $B = 1.54 \times 10^5$ Mc/sec; j is the quantum number for the total angular momentum of the lower rotational level.

Levels with $j \sim 10$ are fairly well populated at room temperature. Consequently, in accordance with Eq. (2.1), ammonia gas at room temperature exhibits rather intense lines at wave lengths λ of the order of several hundred millimeters.

Each rotational level is split into two inversion sublevels. The wavelength corresponding to the transition between inversion levels depends on the quantum numbers j and k (k is the projection of j on the symmetry axis of the molecule) and lies between 1 and 2 cm. If a beam of these molecules passes through a quadrupole focuser, the beam at the output of the focuser contains molecules in the upper inversion levels.

The inversion state of the molecule changes for dipole transitions between rotational levels,¹⁴ so that a beam of molecules selected in this way is characterized by a negative temperature not only for transitions between neighboring inversion levels, but also for rotational-inversion transitions. A quadrupole focuser can also be used to select other molecules¹⁵ (HDO , CH_2O).

A major shortcoming of this method is the fact that only a relatively small number of active molecules* can be obtained. Molecular beam sources presently available are capable of producing beams with maximum fluxes of 10^{15} particle/cm²·sec in each of the rotational levels.

3. Excitation of Gas Molecules by Means of a Gas Discharge

It has been recently pointed out^{16,17} that negative temperatures can be produced in a gas discharge by electronic excitations of the molecules or atoms of the gas. In the work referred to, the stationary processes characteristic of the passage of an electron beam through a gas are considered. A fixed electric field F is applied to the system (Fig. 3).

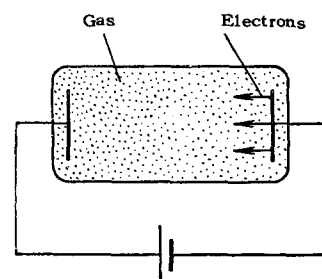


FIG. 3. Arrangement for exciting gas molecules with an electron beam.

Elastic collisions of electrons with the gas atoms in the presence of the external electric field F result in electron "heating", i.e., the mean kinetic energy of the electron can be appreciably greater than the mean kinetic energy of the gas atoms. Inelastic collisions of the first kind between electrons and atoms cause excitation of the atoms, whereas inelas-

*The number of active molecules is equal to the difference between the number of molecules in the upper and lower energy levels.

tic collisions of the second kind cause the atoms to make transitions from higher to lower energy levels.

The number of atomic transitions per unit time from a state with energy ϵ_1 to a state with energy ϵ_2 ($\epsilon_2 > \epsilon_1$) due to collisions of the first kind is

$$\frac{1}{\theta_{12}} = \int_{\epsilon_2 - \epsilon_1}^{\infty} \sigma_{12}(\epsilon) v dn(\epsilon), \quad (3.1)$$

where θ_{12} is the lifetime for a transition from level 1 to level 2, σ_{12} is the cross section for a collision of the first kind between an electron with energy ϵ and an atom, v is the relative velocity of the electron and the atom, (v is actually the electron velocity) and $dn(\epsilon)$ is the number of electrons per cubic centimeter with energies between ϵ and $\epsilon + d\epsilon$.

Similarly, the number of transitions in the reverse direction, from ϵ_2 to ϵ_1 , is

$$\frac{1}{\theta_{21}} = \int_0^{\infty} \sigma_{21}(\epsilon) v dn(\epsilon), \quad (3.2)$$

where σ_{21} is the cross section for collisions of the second kind. The limits of integration are different in Eqs. (3.1) and (3.2): in Eq. (3.1) the integration is taken from some initial energy $\epsilon_2 - \epsilon_1$, which represents the minimum electron energy required for excitation of the atom.

For simplicity we assume that the electronic energy distribution is Maxwellian, with a temperature T_e ; as indicated above, $T_e \gg T_a$, where T_a is the temperature of the gas atoms. Then, in Eqs. (3.1) and (3.2) we have

$$dn(\epsilon) = a e^{-\epsilon/kT_e} e^{1/2} d\epsilon. \quad (3.3)$$

Using the principle of detailed balancing¹⁸ we can now write

$$\epsilon_1 \sigma_{12}(\epsilon_1) = \epsilon_2 \sigma_{21}(\epsilon_2). \quad (3.4)$$

From (3.1), (3.2), and (3.4) it follows that

$$\frac{\theta_{21}}{\theta_{12}} = e^{-\frac{\epsilon_2 - \epsilon_1}{kT_e}}. \quad (3.5)$$

Equation (3.5) indicates that the lifetime in the higher energy state is somewhat shorter than in the lower state if the atomic transitions are due solely to collisions with electrons.

The change in the number of atoms N_1 and N_2 in levels ϵ_1 and ϵ_2 per unit time, due to collisions of the first and second kind, is

$$\frac{dN_1}{dt} = -\frac{dN_2}{dt} = -\frac{N_1}{\theta_{12}} + \frac{N_2}{\theta_{21}}, \quad (3.6)$$

where N_2 is the number of atoms in the ϵ_2 level. In the stationary state $dN/dt = 0$, and from Eq. (3.5) it follows that

$$\frac{N_2}{N_1} = e^{-\frac{\epsilon_2 - \epsilon_1}{kT_e}}. \quad (3.7)$$

Thus, the distribution of atoms over levels is a Boltzmann distribution with a temperature equal to the elec-

tron temperature T_e . Taking account of the degeneracy of the atomic levels we obtain an obvious extension of Eq. (3.7):

$$\frac{N_2}{N_1} = \frac{g_2}{g_1} e^{-\frac{\epsilon_2 - \epsilon_1}{kT_e}}, \quad (3.8)$$

where g_i is the degeneracy of the i -th level.

When the atomic transitions from higher energy levels to lower levels are not due exclusively to collisions of the second kind, but to other processes as well, for example, radiative decay, the distribution over atomic levels differs from that given in (3.7) and (3.8).

In this case the stationary distribution can be obtained by analysis of the change in the population of a given level. If the lifetime for radiative decay in the i -th level is τ_i , then

$$\frac{dN_2}{dt} = \frac{N_1}{\theta_{12}} - \frac{N_2}{\theta_{21}} - \frac{N_2}{\tau_2}. \quad (3.9)$$

If $\tau_2 \ll \theta_{21}$, then in the stationary state we have

$$\frac{N_2}{N_1} = \frac{\tau_2}{\theta_{21}} e^{-\frac{\epsilon_2 - \epsilon_1}{kT_e}}. \quad (3.10)$$

Comparison of (3.7) and (3.10) shows that the distribution of atoms over energy levels differs from the Boltzmann distribution because of the factor $\tau_2/\theta_{21} \ll 1$.

If we introduce an additional energy level, to be definite, say a higher level, it follows from (3.10) that

$$\frac{N_3}{N_2} = \frac{\tau_3}{\theta_{31}} \frac{\theta_{21}}{\tau_2} e^{-\frac{\epsilon_3 - \epsilon_2}{kT_e}}. \quad (3.11)$$

In Eq. (3.11) we find the Boltzmann factor

$\exp \left\{ -\frac{\epsilon_3 - \epsilon_2}{kT_e} \right\} < 1$ since $\epsilon_3 > \epsilon_2$; however, the factor in front of the exponential $\frac{\tau_3 \theta_{21}}{\theta_{31} \tau_2}$ can be greater than unity for certain values of the parameters, and the ratio N_3/N_2 can be greater than unity; this case is equivalent to the existence of a negative temperature with respect to the levels designated 3 and 2.

It is apparent from the preceding discussion that negative temperatures are produced if $\tau_1 < \theta_{11}$; this situation sets a limit on the electron density, since θ_{11}^{-1} is proportional to the density of the electron gas.

The oscillation conditions in a system depend crucially on the number of active atoms $N_3 - N_2$. With all other conditions being equal, it is desirable to have a high density, i.e., the pressure in the system should be high. At high densities the absorption of photons produced by spontaneous transitions in other atoms becomes important. This effect tends to increase the lifetime of the atom in the excited state. The effect is useful when it increases the lifetime of the atom in the upper level 3, that is to say, the upper level must be optically connected with the ground state (allowed transition). Under these conditions the system can operate at higher densities. However, if the ground state is

optically connected to the lower negative temperature level 2, a high density is undesirable, since it increases the relative population of level 2.

In reference 16 neon is considered as an example. The upper level 3 and the lower level 2, between which a negative temperature state is maintained, are the $2s_4$ and $2p_{10}$ levels. The $2p_{10}$ level decays to a lower level in a time $\tau_2 \sim 10^{-8}$ sec. At a pressure of approximately 10 mm Hg the lifetime of the upper level $2s_4$ is estimated to be $\tau_3 \approx 60 \tau_2$. In order to obtain $N_3 = 10^{10} \text{ cm}^{-3}$ at an electron gas temperature $kT_e = 2.9 \text{ eV}$, we must take $\tau_3/\theta_{31} = 3 \times 10^{-5}$. The exact value of the cross section for excitation of the atom to the $2s_4$ level by an electron is not known; however, if we assume that this cross section is $\sigma \approx 10^{-17} \text{ cm}^2$, the electron density in the beam is found to be $n \sim 5 \times 10^{10} \text{ cm}^{-3}$. These values of density and temperature are reasonable for electron beams that can be achieved in practice.

The ratio τ_i/θ_{i1} is essentially the same for all levels i with optically allowed transitions since τ_i^{-1} and θ_{i1}^{-1} are each proportional to the square of the dipole moment matrix element for the transition being considered.^{19,20} As can be seen from Eq. (3.11), negative temperatures cannot be obtained in this case. If both levels are optically connected to the ground state, optical transitions between the levels themselves are forbidden by the selection rules (in the first approximation) and these levels cannot be used in practice for obtaining radiation.

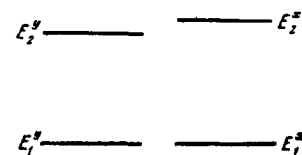
At least one of the two levels in the system must thus be associated with an optically forbidden transition. This level is excited in electron collisions as a result of exchange effects between the incident electrons and the electrons in the atom. The excitation cross section for this process is generally smaller than for optically allowed transitions.

In the example considered above, the lower level in the negative-temperature system is associated with an optically forbidden transition. The small excitation cross section for this level (by electron collisions) favors the production of a negative temperature. However, in such cases, the level being considered may be optically connected with some other excited metastable level of the atom. The existence of a large number of such metastable levels and a high cross section for excitation by electrons to the level in question can increase the population of the lower level. This effect is enhanced by possible capture by the metastable atom of a photon which is produced in emission from the level being considered.

This situation does not arise in neon. In reference 16 it is proposed to reduce the concentration of metastable neon atoms by the addition of a small amount of a quenching gas, for example, argon.

Another method of obtaining negative temperatures is based on the interaction between atoms of two different gases (Fig. 4).

FIG. 4. Level diagram for two interacting gases: x) operating gas, y) quenching gas.



If the energies of any two excited atomic levels in the two gases differ by a small amount Δ , the cross sections for inelastic collisions between these atoms can be appreciable. Under these conditions an atom can be excited by colliding with another atom as well as by collisions with electrons. In interatomic collisions the atoms exchange excitation energy. If this effect is to disturb the equilibrium distribution (3.7), obviously the probability of excitation transfer by the atoms must exceed the probability $1/\theta_{0i}$ of electronic excitation of the atom to a given level.

In this approximation we can obtain the equilibrium distribution of atoms over excited energy levels. For simplicity, let us assume that the atomic energy distribution is Maxwellian with a temperature T :

$$dn(\epsilon) = a e^{-\frac{\epsilon}{kT}} \sqrt{\epsilon} d\epsilon, \quad (3.12)$$

where $\epsilon = p^2/2M$ is the kinetic energy of the atom; the constant a is determined from the normalization condition

$$\int_0^{\infty} dn = n, \quad (3.13)$$

and n is the number of gas atoms in the given excited state per cubic centimeter.

Let us assume that the excitation energy of gas x , which we will denote by E_2^x , is greater than the excitation energy of gas y , denoted by E_2^y , i.e., $E_2^x - E_2^y = \Delta$. Then the probability of excitation of x atoms by y atoms is

$$W_{xy} = a_y^y \int_{\Delta}^{\infty} e^{-\frac{\epsilon}{kT}} \sigma_{xy} v \sqrt{\epsilon} d\epsilon. \quad (3.14)$$

Here, the constant a is determined from the condition

$$a_y^y \int_0^{\infty} \sqrt{\epsilon} e^{-\frac{\epsilon}{kT}} d\epsilon = n_y^y, \quad (3.15)$$

where n_y^y is the number of excited y atoms per cubic centimeter, σ_{xy} is the cross section for transfer of excitation from a y atom to an x atom, and v is the relative velocity of the atoms.

For the inverse process

$$W_{yx} = a_y^x \int_0^{\infty} \sigma_{yx} v e^{-\frac{\epsilon}{kT}} \sqrt{\epsilon} d\epsilon, \quad (3.16)$$

where σ_{yx} is the cross section for the inverse process and a_y^x , as in Eq. (3.15), is determined by the density of y atoms in the ground state 1.

Applying the principle of detailed balancing, as in (3.4), we have from Eqs. (3.14) and (3.16)

$$\frac{W_{xy}}{W_{yx}} = \frac{n_y^y}{n_1^y} e^{-\frac{\Delta}{kT}}. \quad (3.17)$$

The rate of change of the number of x atoms in the excited state with an energy E_2^x is found from Eqs. (3.14) and (3.16):

$$\frac{dn_2^x}{dt} = W_{xy}n_1^x - W_{yx}n_2^x. \quad (3.18)$$

In the stationary state $dn_2^x/dt = 0$; when (3.17) is substituted in (3.18) the latter assumes the form

$$\frac{n_2^x}{n_1^x} = \frac{n_2^y}{n_1^y} e^{-\frac{E_2^x - E_2^y}{kT}}. \quad (3.19)$$

If the atoms are excited by electron impact only, then obviously we write in place of Eq. (3.19)

$$\frac{n_2^y}{n_1^x} = \frac{n_2^y}{n_1^y} e^{-\frac{E_2^x - E_2^y}{kT_e}}. \quad (3.20)$$

Thus, collision transitions between close-lying levels in different atoms cause a departure from the distribution produced by electronic excitation given in (3.20). Under these conditions the probability of finding an atom in the upper of the two levels being considered in two different atoms is smaller than the value given by (3.20), whereas the probability of finding an atom in the lower level is larger. Hence, it is possible to obtain negative temperatures in the gas between levels whose distribution is given by (3.20) and levels subject to an excitation mechanism such as that described above for collisions between different atoms, whose distribution is given by (3.19).

A shortcoming of this method of obtaining negative temperatures is that strong transfer of excitation in collisions between two kinds of atoms requires that the levels be close together, in which case the effect of redistribution of atoms over the levels is small.

A mixture of Hg at a pressure of 10^{-3} mm Hg and Kr at a pressure of 10–50 mm Hg is analyzed in reference 16. The mercury 9^1P and 6^1F levels lie close to the $5S_5$ metastable level of krypton so that populations of the 9^1P and 6^1F levels can be increased. Under these conditions a negative temperature is to be expected with respect to the 6^1D level.

The metastable $3S_1$ level in He also lies close to the $2s_4$ level in Ne considered above. This situation can also be exploited to increase the population of the $2s_4$ level, thereby providing a negative temperature in the Ne system considered above.

A similar situation prevails when two or more levels in the same atom lie close together in energy. The $2s_2$, $2s_3$, $2s_4$ and $2s_5$ levels in Ne have approximately the same energy and the population of the lower of the two $2s_5$ levels could be increased greatly, thereby producing a negative temperature with respect to the $2p_{10}$ level.

The relatively low density of excited atoms is a great disadvantage in both of these methods of obtaining negative temperatures in a gas by means of electron beams.

It should be noted, however, that, in principle, there is no limitation in this method on the frequencies corresponding to transitions between the negative temperature levels. A somewhat different method of obtaining negative temperatures has been analyzed and studied experimentally in reference 79.

A glow discharge was used to excite mercury vapor to which hydrogen had been added. The hydrogen strongly perturbs the mercury atoms in the 6^3P level; the $7^3S_1 \rightarrow 6^3P_{0,1,2}$ transition is then used to obtain a negative temperature. Another mercury tube was used to investigate the transmission through the discharge tube containing the mixture of mercury and hydrogen vapors. The results of the experiments indicate that in certain cases the transmission factor can be greater than unity. The authors point out that the theoretical interpretation of the experiment is difficult because of the lack of reliable data for discharges of this kind.

4. Pulse Method of Obtaining Negative Temperatures in Semiconductors*

A great deal of data is available concerning the optical properties of semiconductors.²¹ These data are well explained by the band theory of solids.^{21,22} Electromagnetic absorption spectra in semiconductors are due basically to the following: 1) electron transitions from valence bands to conduction bands; 2) transitions from impurity levels into bands, 3) internal band absorption, associated with the presence of free carriers; 4) absorption by the crystal lattice.

In this section we consider the possibilities of obtaining negative temperatures between bands and between impurity levels and associated bands which have been proposed in reference 23.

According to the band theory of solids, a semiconductor can be described by a series of energy bands which represent forbidden and allowed electron states. In a pure semiconductor there are no electrons in the upper energy band (conduction band) at $T \rightarrow 0$ while the energy states in the lowest band (valence band) are completely filled by electrons. If a semiconductor contains impurities, these impurities form additional levels which, as a rule, are located in the forbidden band. There are two kinds of impurities in a semiconductor: impurities that can give electrons to the conduction band (donors) and impurities that can trap electrons from the valence band (giving rise to conduction by holes), called acceptors. The distribution of electrons over energy levels in a semiconductor at thermodynamic equilibrium is described by the Fermi distribution function

$$f(\epsilon) = (e^{\frac{\epsilon - \mu}{kT}} + 1)^{-1}, \quad (4.1)$$

*The possibility of obtaining negative temperatures in semiconductors between cyclotron-resonance levels and between levels in impurity atoms is considered in reference 80, which appeared while the present paper was in press.

where the chemical potential μ is determined from the normalization condition

$$\int f(\epsilon) d\Gamma = N, \quad (4.2)$$

where N is the total number of electrons. For electrons in the conduction band*

$$d\Gamma = \rho(\epsilon) d\epsilon, \quad \rho(\epsilon) = \frac{V \sqrt{2} m^{3/2} e^{3/2} V}{\pi^2 \hbar^3}. \quad (4.3)$$

For discrete spectra the integral in Eq. (4.2) is replaced by a summation over quantum-mechanical states.

A negative temperature can be obtained in a semiconductor by ionizing the impurities or the valence band by a pulsed electric field. The peak value of the pulse is chosen to cause impact ionization of the valence band or the Zener effect. The number of electrons in the conduction band then increases sharply. The number of electrons ejected into the band is determined by the breakdown conditions (the strength of the electric field and the length of the pulse).

If the pulse decays rapidly enough, all the electrons are transferred to the lowest energy levels of the associated band if their lifetime in the band is long enough. The electron density and crystal temperature must be chosen to obtain a state which is more or less degenerate; this is equivalent to a negative temperature with respect to some narrow range of free higher energy levels in the valence band. In accordance with Eq. (1.3), for a negative temperature, the condition $f(\epsilon) \geq 0.5$ must obtain for some region close to the bottom of the conduction band and $f(\epsilon) \leq 0.5$ for some region close to the top of the valence band (Fig. 5).

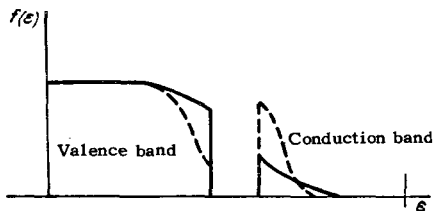


FIG. 5. Energy distribution of electrons in a semiconductor. The solid line is the thermodynamic equilibrium distribution. The dashed line is the nonequilibrium distribution corresponding to a negative temperature between the valence band and the conduction band.

The large electron densities required for degeneracy mean that rather high electric fields must be applied to the sample. A number of theoretical and experimental papers²⁴⁻²⁹ treat problems connected with electrical breakdown in semiconductors; in the present review, breakdown in semiconductors will not be considered in detail. It should be indicated, however, that according to the work cited above, breakdown of valence semiconductors takes place in fields in which the mean energy of the conduction electrons is of the

*All the considerations given here also apply for holes.

same order of magnitude as the ionization potential. The field F_i at which the mean electron energy is of the same order as the ionization potential $\epsilon \approx \epsilon_i$ is determined from the condition

$$F_i \approx \frac{(\hbar\omega_0 m)^{1/2}}{e\tau_{0n}(\epsilon_i)}, \quad (4.4)$$

where m is the effective mass of the carrier, ω_0 is the frequency of the optical vibrations of the lattice, τ_{0n} is the relaxation time of the carrier with respect to optical vibrations of the lattice [cf. Eq. (4.16)].

The energy distribution of conduction electrons in a strong electric field, which takes account of collisions with lattice vibrations,²⁷ is of the form (Fig. 6)

$$f(\epsilon) = N' \exp \left\{ -\frac{\epsilon^2}{2p'(kT)^2} \right\}, \quad (4.5)$$

where

$$p' = \frac{(eFl_a)^2}{6mu^2kT} (1+R)^{-1} + R \frac{(\hbar\omega_0)^2}{mu^2kT},$$

$$R \approx \frac{D^2}{C^2}, \quad l_a = \frac{9\pi Mu^2 \hbar^4 n_0}{4C^2 m^2 kT}.$$

Here F is the applied electric field, u is the velocity of sound in the semiconductor, M is the mass of the lattice atom, n_0 is the density of atoms in the lattice,

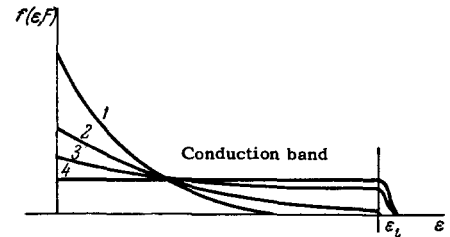


FIG. 6. Energy distribution of electrons in a conduction band as a function of the strength of the applied electric field. The numbers on the curves increase with increasing field strength. Beyond the ionization threshold (ϵ_i) the distribution function falls off exponentially.

k is the Boltzmann constant, T is the lattice temperature, and C^2 and D^2 are interaction constants for the acoustic and optical lattice vibrations respectively.

The energy distribution of the electrons in the conduction band is approximately a Boltzmann distribution with an effective temperature $T_e \gg T$.

When the electric field is switched off two processes take place in the semiconductor: 1) nonequilibrium carriers recombine, and 2) the temperature of the nonequilibrium carriers is reduced to the temperature of the crystal. Negative temperatures can obtain in a semiconductor only when the characteristic time for the second process is much smaller than the lifetime of the nonequilibrium carriers τ_c . At the present time there is no general theory which allows us to determine the time τ_c . There are a number of experimental methods of determining τ_c , however, but these give inconsistent values.^{74,75} As a rule, τ_c is determined by nonradiative transitions and is associated with the

presence of impurities and defects in the lattice.

A theoretical analysis of the time t during which a negative temperature obtains (the thermalization time of nonequilibrium carriers) in semiconductors is given in reference 30.

By nonequilibrium (fast) carriers we mean electrons* (holes) with an initial energy ϵ_0 which is appreciably higher than the mean thermal energy or degeneration energy (if the gas is degenerate), but which does not exceed the threshold for collision ionization of the valence band ϵ_i .

Electron thermalization in crystals is due primarily to scattering on lattice vibrations. Impurities do not play an important role in slowing down electrons because the energy lost by an electron in collision with an impurity atom (of order m/M , where M is the mass of the impurity atom) is much smaller than the energy lost in emission of a phonon, i.e., small compared with the energy lost in lattice collisions.

Electron thermalization can be analyzed by considering the kinetic equation for the electron momentum distribution in the conduction band of the crystal $f(\mathbf{p})$. In the kinetic equation we must take account of the Fermi degeneracy of the electron gas since this factor becomes important in the final stages of thermalization, at which point the quantum states are almost completely filled. The electron-electron collision term can usually be neglected since it does not contribute directly to the reduction of the mean electron energy. However, at high electron densities electron-electron collisions can have an important effect on the form of the distribution function so that this effect will be taken into account.

For simplicity we shall assume an isotropic dispersion relation $\epsilon = p^2/2m$. Since there is no external field, the distribution function depends only on the electron energy, i.e., $f(\mathbf{p}) = f(\epsilon)$.

We expand the collision integral by the usual method,²⁸ in powers of the small quantity $\hbar\omega_{\mathbf{q}}/\epsilon(\mathbf{p})$, where $\hbar\omega_{\mathbf{q}}$ is the energy of a phonon with momentum \mathbf{q} ; this procedure yields the following equation for $f(\epsilon)$:

$$\frac{\partial f(\epsilon)}{\partial t} = \frac{1}{p} \frac{\partial}{\partial \epsilon} \left\{ G(\epsilon) \left[f(\epsilon) (1 - f(\epsilon)) + \eta(\epsilon) \frac{\partial f(\epsilon)}{\partial \epsilon} \right] \right\}, \quad (4.6)$$

where

$$G(\epsilon) = \frac{Vm}{2\pi\hbar^4} \int_0^{2p} B(q) \hbar\omega_{\mathbf{q}} q dq,$$

$$G(\epsilon) \eta(\epsilon) = \frac{Vm}{4\pi\hbar^4} \int_0^{2p} B(q) (\hbar\omega_{\mathbf{q}})^2 (2n_{\mathbf{q}} + 1) q dq,$$

where $B(q)$ is the square of the matrix element for the electron-phonon interaction and $n_{\mathbf{q}}$ is the number of phonons with energy $\hbar\omega_{\mathbf{q}}$

*Hereinafter we consider only electrons in the conduction band, although the results also apply for holes in the valence band.

$$n_{\mathbf{q}} = (e^{\frac{\hbar\omega_{\mathbf{q}}}{kT}} - 1)^{-1}, \quad (4.7)$$

and V is the volume of the crystal.

The electron-lattice collision time τ is

$$\tau^{-1} = \frac{v}{l} = \frac{Vm}{4\pi\hbar^4 p^3} \int_0^{2p} B(q) (2n_{\mathbf{q}} + 1) q^3 dq, \quad (4.8)$$

where v is the electron velocity and l is the mean free path. We multiply Eq. (4.6) by $\epsilon\rho(\epsilon)$, where the density of states of electrons with energy ϵ is

$$\rho(\epsilon) = \frac{\sqrt{2m^2\epsilon^2V}}{\pi^2\hbar^3},$$

and integrate over the electron energies, thereby obtaining

$$\frac{dE}{dt} = - \int_0^{\infty} \frac{\rho(\epsilon)}{p} G(\epsilon) \left\{ f(\epsilon) [1 - f(\epsilon)] + \eta(\epsilon) \frac{\partial f}{\partial \epsilon} \right\} d\epsilon. \quad (4.9)$$

Here $E = \int_0^{\infty} \epsilon\rho(\epsilon) f(\epsilon) d\epsilon$ is the mean electron energy.

The first term in the integral of Eq. (4.9) describes the loss of electron energy due to spontaneous phonon emission; the second term arises from electron diffusion in energy space. As will be evident below, taking account of the second term in the integral of Eq. (4.9) in scattering of electrons on acoustic lattice vibrations leads to a logarithmic infinity in the time required for establishing equilibrium between the electrons and the lattice. The second term in Eq. (4.9) need not be taken into account in the analysis of thermalization of electrons to an energy which is greater than the lattice temperature.

Let us consider the thermalization of an individual electron and neglect degeneracy. In this case,

$$f(\epsilon) \rho(\epsilon) = \delta(\epsilon - E). \quad (4.10)$$

I. For acoustic phonons³¹

$$G(\epsilon) = \frac{16m^3 C^2 \epsilon^2}{9\pi\hbar^4 M n_0}. \quad (4.11)$$

Using (4.4) and (4.10), we obtain from (4.9)

$$\frac{dE}{dt} = -a_0 E^{\frac{3}{2}}. \quad (4.12)$$

where a_0 is expressed in the following way in terms of the mobility in scattering on acoustic vibrations of the lattice w :

$$a_0 = \frac{8w^2}{3\sqrt{\pi} w(T) (kT)^{\frac{3}{2}}}. \quad (4.13)$$

The time to go from energy E_0 to energy E is

$$t = \frac{2}{a_0} (E^{-\frac{1}{2}} - E_0^{-\frac{1}{2}}). \quad (4.14)$$

It is important that the thermalization time t depends only on two semiconductor constants, the mobility and the velocity of sound. When $E_0 \gg E$, the time to reach an energy corresponding to $T \approx 300^\circ\text{K}$ for

electrons in Ge ($w \approx 3600 \text{ cm}^2 \text{ v}^{-1} \text{ sec}^{-1}$ at room temperature and $u \approx 4.94 \times 10^5 \text{ cm/sec}$) is $t \approx 4.5 \times 10^{-10} \text{ sec}$; for electrons in

Si ($w \approx 1200 \text{ cm}^2 \text{ v}^{-1} \text{ sec}^{-1}$ and $u \approx 8.5 \cdot 10^5 \text{ cm/sec}$),
 $t \approx 5.1 \cdot 10^{-11} \text{ sec}$.

The ratio of the times required to go from energy E_0 to E for $E_0 \gg E$ in two different semiconductors x and y is

$$\frac{t_x}{t_y} = \frac{w_x u_y^2}{w_y u_x^2}. \quad (4.15)$$

Equation (4.12) has a simple physical meaning. The energy lost by the electrons in emission of an acoustic phonon is $\hbar\omega_Q = uq$. Since scattering on acoustic phonons is spherically symmetric, and the maximum momentum of the radiated phonon is twice as large as the electron momentum, $\hbar\omega_Q \sim (2m\epsilon u^2)^{1/2}$. The frequency of collisions is associated with the energy loss and is expressed in terms of the lattice collision time (4.8); we must take account of the fact that there is an energy loss in only one out of $2n_Q + 1$ collisions. Thus,

$$\frac{dE}{dt} \approx -\frac{\hbar\omega_Q}{\tau(2n_Q+1)} \approx -a_0 E^{\frac{3}{2}}.$$

II. Thermalization of fast electrons by optical lattice vibrations in a valence semiconductor is described by Eqs. (4.6) and (4.9) when the electron energy $E \gg \hbar\omega_0$, where $\hbar\omega_0$ is the energy of an optical phonon ($\hbar\omega_0 \approx kT_D$, T_D is the Debye temperature).

In this case³¹

$$G(\epsilon) = \frac{D^2 K^2 m^2 \epsilon}{\pi \hbar^2 M n_0}, \quad \tau^{-1} = \frac{D^2 K^2 m^2 v}{2\pi \hbar^4 M n_0 \hbar \omega_0}, \quad \frac{dE}{dt} = -\frac{K^2 D^2 m^{\frac{3}{2}} E^{\frac{1}{2}}}{\sqrt{2\pi n_0 M \hbar^2}}, \quad (4.16)$$

where K is the reciprocal lattice vector and D^2 is the optical phonon interaction constant, which is analogous to C^2 .

The time to go from energy E_0 to E is

$$t_{\text{opt}} = \frac{2\sqrt{2}\pi M n_0 \hbar^2}{K^2 D^2 m^{\frac{3}{2}}} (E_0^{\frac{1}{2}} - E^{\frac{1}{2}}). \quad (4.17)$$

Taking $C^2 \approx D^2$ and comparing the thermalization times for optical and acoustic phonons, we find

$$\frac{t_{\text{opt}}}{t_{\text{acous}}} \approx \frac{mu^2}{kT_D} \approx 10^{-2}.$$

A negative temperature can be produced in a semiconductor only when the electrons in the conduction band and the holes in the valence band are highly degenerate and are thermalized by the crystal lattice. The degree of degeneracy depends on the temperature (mean energy) of the electron gas and its density. Reduction of the mean energy of the electron gas causes a marked increase in thermalization time so that electrons can recombine before the negative temperature is produced. For this reason it is desirable to increase the density rather than to reduce the en-

ergy.* It is pointed out in reference 24 that energy exchange is stronger in electron-electron collisions than in collisions with acoustic phonons even at electron densities of $n \sim 10^{14} \text{ cm}^{-3}$ (for $T < T_D$ and $\epsilon > kT_D$ the interaction with optical phonons can be neglected).

Thus, at the electron densities necessary for obtaining negative temperatures, the form of the distribution function may be assumed known and taken as:

$$f(\epsilon) = \left(e^{\frac{\epsilon - \mu}{\hbar\theta}} + 1 \right)^{-1}, \quad (4.18)$$

where μ is the chemical potential and θ is the temperature of the electron gas.

The thermalization process means essentially that the temperature θ is reduced to the temperature of the lattice T . We have already determined the thermalization time for electrons without taking account of degeneracy or the second term on the right-hand side of Eq. (4.9). Now, using Eq. (4.9) we can determine the time for the temperature of the electron gas to change from θ_0 to θ , taking account only of the interaction with acoustic phonons.

Introducing Eq. (4.18) in the usual way, we have from Eq. (4.9)

$$\frac{dE}{dt} = -2a_0 (\theta - T) E^{\frac{1}{2}}, \quad (4.19)$$

where

$$E^{\frac{1}{2}} = \int_0^\infty \epsilon^{\frac{1}{2}} Q(\epsilon) f(\epsilon) d\epsilon.$$

Equation (4.19) is a differential equation for $\theta(t)$ which reduces to integration of elementary functions for sufficiently strong degeneration, i.e., when $\mu/k\theta_0 \gg 1$. Under these conditions, the usual procedure for expanding the integrals³² for E , $E^{1/2}$ and μ yields

$$\theta \frac{d\theta}{dt} = -\frac{3a_0}{k\pi^2 \mu_0^{\frac{2}{3}}} (\theta - T) \left[\mu_0^2 + \frac{\pi^2 k^2 \theta^2}{6} \right], \quad (4.20)$$

where

$$\mu_0 = \frac{(\pi \hbar)^2}{2m} \left(\frac{3n}{\pi} \right)^{\frac{2}{3}},$$

and n is the electron density.

Integrating Eq. (4.20) we have

$$t = \frac{k\pi^2}{3a_0 \mu_0^{\frac{2}{3}}} \frac{1}{\left(1 + \frac{\pi^2 k^2}{6\mu_0^2} T^2 \right)} \left\{ T \ln \frac{(\theta_0 - T) \left(1 + \frac{\pi^2 k^2}{6\mu_0^2} \theta^2 \right)^{\frac{1}{2}}}{(\theta - T) \left(1 + \frac{\pi^2 k^2}{6\mu_0^2} \theta_0^2 \right)^{\frac{1}{2}}} + \frac{\sqrt{6}\mu_0}{\pi k} \left[\tan^{-1} \frac{\pi k}{\sqrt{6}\mu_0} \theta_0 - \tan^{-1} \frac{\pi k}{\sqrt{6}\mu_0} \theta \right] \right\}. \quad (4.21)$$

When $\theta \rightarrow T$, the first term in the brackets in Eq. (4.21), which derives from the second term on the

*The electron density, however, must not be high enough to give a negative dielectric constant in the frequency region of interest.

right-hand side of Eq. (4.9), approaches infinity in logarithmic fashion, corresponding to an infinite time for establishment of equilibrium between the electrons and the lattice. When $\theta > T$, the logarithm in Eq. (4.21) can be neglected and the thermalization time is given by the following expression:

$$t \approx \frac{k\pi^2}{3a_0\mu_0^2} (\theta_0 - \theta). \quad (4.22)$$

To compare the thermalization time with and without degeneracy [cf. Eq. (4.14)], it is convenient to replace the temperature θ by the mean electron energy $\bar{\epsilon}$ in Eq. (4.22). Then ($\theta_0 \gg \theta$)

$$t \approx \frac{2}{a_0 e^2} \frac{\pi}{5} \left(\frac{\epsilon_0}{\bar{\epsilon}} - 1 \right)^{\frac{1}{2}},$$

where

$$\epsilon \approx \frac{3}{5} \mu_0, \quad \epsilon_0 \approx \frac{3}{5} \mu_0 \left(1 + \frac{5\pi^2 k^2 \theta_0^2}{12\mu_0^2} \right).$$

Although Eqs. (4.14) and (4.22), have different forms, both give thermalization times of the same order of magnitude.

Thermalization of a degenerate electron gas on optical phonons can be neglected because in cases of practical interest the mean electron energy is less than $\hbar\omega_0$ when degeneracy is important. Thus, the thermalization of the electron gas in a semiconductor can be divided into two stages: 1) thermalization on optical vibrations of the lattice to an energy of approximately $\hbar\omega_0$, with a short characteristic time (4.17), and 2) a further thermalization on acoustic vibrations with a much longer characteristic time, as described by (4.14) and (4.21).

Thus, the time during which a negative temperature obtains is determined basically by thermalization of the electrons from an energy $\hbar\omega_0$ to an energy corresponding to the required degree of degeneracy, with the sole mechanism being the interaction with acoustic lattice vibrations.

We now consider in greater detail the pulse method for producing negative temperatures between impurity levels and the associated band in a semiconductor. For impact ionization, the field for which there are $N-n$ electrons in the impurity levels in the stationary state is found from the following condition:

$$\alpha n(N-n) - \beta n^2 + g_T(N-n) = 0, \quad (4.23)$$

where β is the probability of recombination of conduction electrons with the impurity levels, α is the collision ionization coefficient (field dependent), N is the number of donor impurities per cubic centimeter, n is the number of electrons in the conduction band and g_T is the probability of thermal ejection into the conduction band (per second).

Direct ionization of impurities by the field (Zener effect) is described by the equation,

$$\gamma(N-n) - \beta n^2 + g_T(N-n) = 0, \quad (4.24)$$

where γ is the probability of ionization of an impurity atom by the field (per second).

Thermal ejection can be neglected at low temperatures in Eqs. (4.23) and (4.24). Then, if the number of electrons in the impurity levels is to be smaller than ΔN ($\Delta N \ll N$), when Eq. (4.23) holds, we require that

$$\alpha > \left(\frac{N}{\Delta N} \right) \beta; \quad (4.25)$$

when Eq. (4.24) holds, this requirement becomes

$$\gamma > \frac{N}{\Delta N} \beta N. \quad (4.26)$$

The impurity levels are emptied by the mechanism which first satisfies one of the inequalities given above at low fields.

If α satisfies the condition in (4.25) for some field while γ has not reached the value αN , the impurity population is reduced to ΔN by impact ionization, and vice versa. From a knowledge of the distribution function for the conduction electrons in a strong field we can obtain the field dependence of the collision ionization coefficient if ionization is treated as a perturbation. When scattering on optical lattice vibrations can be neglected (low temperatures and shallow impurities), α is given by:³³

$$\alpha(F) = \frac{5\sigma_0 \left(\frac{I}{m} \right)^{\frac{1}{2}} \left(\frac{F}{F_0} \right)^4 e^{-\frac{F_0^2}{F^2}}}{\Gamma\left(\frac{3}{5}\right) \left(\frac{F}{F_0} \right)^5}, \quad (4.27)$$

where $\Gamma(x)$ is the gamma function, I is the impurity ionization potential, and σ_0 is the maximum ionization cross section

$$F_0^2 = \frac{24}{25} \sqrt{\frac{I}{mu^2}} \left(\frac{2Imu^2}{ekTl} \right)^2, \quad F_0^{*2} = \frac{5}{2} F_0^2. \quad (4.28)$$

The probability of ionization of an impurity atom by the field can be estimated if we know the probability for ionization of the ground state of the hydrogen atom; we change the ionization potential in the appropriate formula and replace the ordinary mass by the effective mass. We have³⁴

$$\gamma(F) = \frac{16I^2 em}{F\hbar^3} e^{-\frac{4(2m)^{1/2} I^{3/2}}{3e\hbar F}}. \quad (4.29)$$

The expression (4.29) for $\gamma(F)$, shows that with a sufficiently strong field we can always satisfy the condition in (4.26), i.e., the required emptying can always be achieved by virtue of the Zener effect. The situation is somewhat different with Eq. (4.25) for α . The exact dependence of α on field intensity F cannot be determined for very strong fields. Hence, the condition in (4.25) will not be satisfied if the coefficient β is very large. However, β is probably small because in a strong field electrons cannot recombine via the transfer of only a single phonon. On the other hand, the quantity g_T in Eq. (4.23) can be large in a strong field because of the Frenkel effect.³⁵ The last two mechanisms can provide the required emptying even

before the Zener effect. Estimates show that marked emptying of shallow impurities (for example Sb in germanium) occurs at fields of 20 v/cm.

As has been indicated above, the production of a negative temperature between impurity levels and the associated band is possible so long as the lifetime of the excited carriers τ_c is greater than the thermalization time t , which is actually determined by Eq. (4.14), i.e.,

$$\tau_c > \frac{2}{a_0 (kT)^{\frac{1}{2}}}. \quad (4.30)$$

In Eq. (4.14) it is assumed that $E_0 \gg E$ while $E \approx kT$, where T is the temperature of the semiconductor lattice. On the other hand, for impurity recombination τ_c is given by the expression

$$\tau_c^{-1} \approx v\sigma_c (N - n), \quad (4.31)$$

where v is the electron velocity and σ_c is the cross section for nonradiative recombination.

Since the existence of a negative temperature requires that there be degeneracy in the conduction band, the temperature of the crystal must satisfy the familiar relation

$$T < 2\pi m^{-1} k^{-1} \hbar^2 n^{\frac{2}{3}}. \quad (4.32)$$

From (4.30), (4.31), and (4.32), it follows that

$$T < 2\pi k^{-1} \hbar^2 \left(\frac{a_0}{m\sigma_c} \right)^{\frac{2}{3}}. \quad (4.33)$$

For the impurity³⁶ Sb in Ge ($\sigma_c \approx 5 \times 10^{-13} \text{ cm}^2$, $m \approx 2.5 \times 10^{-28} \text{ g}$), we find from Eq. (4.33) $T < 1.3^\circ \text{K}$.

The impurity concentration at the required temperature is found from the condition in (4.32)

$$n = \left(\frac{kmT}{2\pi\hbar^2} \right)^{\frac{3}{2}}. \quad (4.34)$$

An estimate for electronic Ge at $T \approx 1^\circ \text{K}$ gives $n \approx 4.2 \times 10^{14} \text{ cm}^{-3}$; thus, we find from (4.31) $\tau_c \approx 0.5 \times 10^{-8} \text{ sec}$.

It should be noted that the oscillation condition (Sec. 8) imposes a limitation on the minimum number of electrons in the conduction band.

The estimates given above show that for Ge and other impurity semiconductors negative temperature states can be obtained in principle. However, the necessity of using low temperatures and of switching the field off rapidly imposes great difficulties.

In interband transitions the electric field required for producing a sufficient concentration of electrons in the conduction band is a sensitive function of the width of the forbidden band. Thus, high intensities and relatively short pulses are required in semiconductors with large forbidden bands. If the effective mass of the carrier in the semiconductor is not small compared with the mass of the electron, the concentrations needed for degeneracy imply high currents and high powers; in this case it is difficult to keep the

sample at a low temperature. This is why it is difficult to obtain negative temperatures in such familiar semiconductors as germanium and silicon. The production of negative temperatures is facilitated through the use of semiconductors with narrow forbidden bands and carriers with small effective mass (for example, InSb) if the lifetime of the nonequilibrium carriers is long enough. It should be noted that the lifetime of nonequilibrium carriers in interband recombination in pure crystals is, as a rule, many times greater than the lifetime of nonequilibrium carriers with respect to impurity levels.

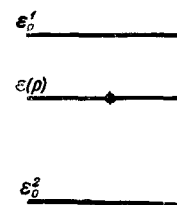
At the present time it is still difficult to choose semiconductor materials suitable for the production of negative temperatures because the characteristics of highly purified semiconductor compounds are not known.

5. Production of Negative Temperatures Between Levels in the Same Band in Semiconductors ("Negative Mass Amplifiers")

In reference 37 the suggestion is made that electromagnetic waves can be amplified by so-called negative-mass carriers in a semiconductor; the mass of these carriers is "negative" because in moving through a field these carriers can transfer their energy to the field, i.e., they exhibit negative losses.

Consider an electron with energy $\epsilon(p)$ (Fig. 7) in a conduction band in a semiconductor.* In interacting with radiation this electron can absorb a photon of energy $\hbar\omega$ and, by this means, be transferred to a higher energy level; this electron can also radiate a photon and drop to a lower energy level. The probability of these two processes depends on the level structure of the band, i.e., the matrix element for the transition and the level density of the final state.

FIG. 7. Energy diagram for a conduction band: $\epsilon(p)$ is the energy of an electron with momentum p , ϵ_0^2 is the lower boundary of the conduction band, ϵ_0^1 is the upper boundary of the conduction band.



When the matrix elements for absorption and emission of photons are the same, as is generally the case, the probability of a transition from the level $\epsilon(p)$ to $\epsilon(p) + \hbar\omega$ or $\epsilon(p) - \hbar\omega$ is determined by the density of final states. Thus, the probability of induced emission of a photon by the electron will be greater than the probability of absorption if the density of final states $\rho(\epsilon - \hbar\omega)$ is greater than $\rho(\epsilon + \hbar\omega)$ and vice versa. When the electron energy covers several states an average must be taken over these states.

Thus, in order for an individual electron to radiate under the effect of an external field, it must be in a state for which

*These considerations also apply for holes in the valence band.

$$\frac{\partial \rho(\epsilon)}{\partial \epsilon} < 0, \tag{5.1}$$

i.e., the density of states must fall off with increasing energy.

For isotropic dispersion we can write $\epsilon(p) = \epsilon_0 + (p - p_0)^2/2m$ near the top and bottom of the band, where the constants ϵ_0 and p_0 are respectively the limiting values of energy and momentum; the sign of $\partial \rho(\epsilon)/\partial \epsilon$ determines the sign of the effective mass of the carrier m , which is given by

$$m^{-1} = \frac{\partial^2 \epsilon(p)}{\partial p^2}. \tag{5.2}$$

It follows from Eq. (5.2) and from the dispersion relation that the effective mass is positive for an electron in levels in the lower part of the conduction band and negative for levels in the upper part of the conduction band. Thus, an electron can radiate by stimulated emission if it is in a negative mass state, i.e., if it is in the upper part of the conduction band.

The situation can be illustrated by a classical analysis of the behavior of an electron in a semiconductor in the presence of an electric field. Suppose that the electron is in a negative mass state with energy $\epsilon(p_1)$. In the electric field F the electron experiences a force $-eF$ (Fig. 8). To be definite we shall assume that the field direction is such that the force which acts on the electron is in the direction of positive momentum p . Because of the field, in a time dt the electron goes from momentum p_1 , to $p_1 + dp$, where $dp = -eF dt$.

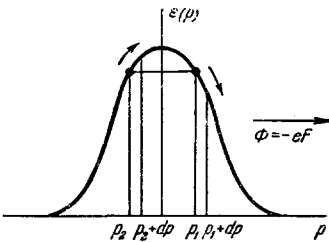


FIG. 8. Diagram of possible transitions for two electrons in a conduction band under the effect of an applied external field; the magnitudes of the momentum are the same but the directions are different.

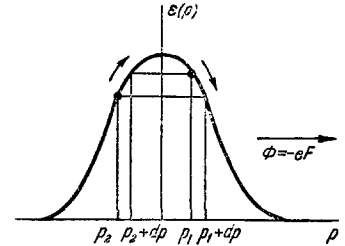
In the new state the electron energy is less than the original energy; the net effect is an energy transfer from the carrier to the electric field, i.e., amplification of the field. However, for an electron which is in an initial state of the same energy ϵ but different momentum p_2 (Fig. 8), so that $\epsilon(p_1) = \epsilon(p_2)$, the transition to the new state $p_2 + dp$ corresponds to an increase in electron energy because of the external field. Thus, in negative mass states we can have both amplification and absorption of the energy of the external electromagnetic field. If we now average over electron states of a given energy, as shown above, the total effect is amplification of the external field applied to the semiconductor, in accordance with the quantum-mechanical analysis given earlier.

It should be noted that the foregoing applies to an individual electron in a negative mass state. In semiconductors, however, there is always a finite number of electrons and the behavior of these electrons in the

field must be analyzed by statistical methods. It is then found that a system in thermodynamic equilibrium can only absorb electromagnetic radiation, regardless of the sign of the effective mass of the carrier.

We now consider two electrons: in a time dt one of these, under the effect of the field, goes from energy ϵ_1 to a higher energy ϵ_2 with absorption of field energy; the other goes in the reverse direction (Fig. 9).

FIG. 9. Diagram showing simultaneous transitions under the effect of an external field with the absorption and emission of energy.



It is clear that the external field will be amplified only if the probability of finding an electron in level ϵ_2 is greater than the probability of finding an electron in level ϵ_1 . In thermodynamic equilibrium the level population falls off with energy so that amplification is impossible.

In order to obtain amplification by means of negative mass carriers, as in other cases, thermodynamic equilibrium must be disturbed and the system must be in a state in which the number of transitions from higher levels to lower levels is greater than the number of transitions in the opposite direction. As will be shown below, this state is a negative temperature state.

Since the number of electrons in a state with energy ϵ is proportional to the density of levels $\rho(\epsilon)$, the analysis given above for a single electron must be modified to take account of the distribution over levels. The difference between the number of transitions per unit time in which there is absorption and induced emission, under the effect of n photons, is

$$nW \{ \rho(\epsilon) \rho(\epsilon - \hbar\omega) f(\epsilon) [1 - f(\epsilon - \hbar\omega)] - \rho(\epsilon - \hbar\omega) \rho(\epsilon) f(\epsilon - \hbar\omega) [1 - f(\epsilon)] \}, \tag{5.3}$$

where W is a quantity which is proportional to the square of the matrix element for the interaction of a photon with an electron in the semiconductor. It follows from (5.3) that the following relation must hold

$$f(\epsilon) > f(\epsilon - \hbar\omega) \tag{5.4}$$

if the induced emission is to be stronger than the absorption. This relation is in complete agreement with the general requirement on the form of the distribution function for states in which a system can amplify (negative temperature states) given above in Sec. 1.

It should be emphasized that this result is independent of the sign of the effective mass of the carrier.

In reference 37, two methods have been proposed for using carriers with negative effective mass in a cw semiconductor amplifier. Both methods are based

on an energy analysis and it is proposed to obtain negative-loss states by applying a fixed electric field to the semiconductor. This field is intended to serve as a source of energy for amplification.

The condition in (5.4) was not taken into account in reference 37, however, and a number of authors³⁸⁻⁴¹ have shown that the analysis in reference 37 does not actually prove that a negative-mass amplifier is possible. Indeed, a negative-mass semiconductor amplifier or oscillator must be analyzed in terms of the possibility of disturbing thermodynamic equilibrium and obtaining states which satisfy the condition in (5.4). In reference 37 it is proposed to use the stationary state in an isotropic band in a fixed electric field; however, it can be shown by direct calculation that it is impossible to obtain states which satisfy (5.4) in a semiconductor²⁵⁻²⁸ [cf. Eq. (4.5), Sec. 4]. The distribution function is such that $\frac{\partial f(\epsilon F)}{\partial \epsilon} < 0$ up to the large values of the field ($\frac{\partial f(\epsilon F)}{\partial \epsilon} \rightarrow 0$ for $F \rightarrow \infty$). It is also impossible to obtain amplification in very strong fields (scattering of electrons on lattice phonons can be neglected) even if collision ionization and the Zener effect are neglected. In this case, the electron will move periodically between the top and bottom of the allowed band so that electron states with positive and negative mass are equally probable. Thus, for an isotropic effective mass it is impossible even in principle to obtain states with negative losses under stationary conditions; in the foregoing we have also neglected additional difficulties associated with ionization of the valence band and interactions with optical phonons in strong fields.

In the second method it is proposed to use the anisotropic effective mass effect; in this case the mass can be negative for certain direction of the quasi-momentum \mathbf{p} .

For example, the energy structure of the valence band of germanium for heavy holes is of the form shown in Fig. 10. For this level structure a carrier at certain points inside a cone of angle θ has a negative effective mass in the direction of \mathbf{p}_y . This result follows directly from the curves in Fig. 11. In the absence of a field the carriers lie within a circle close to the origin of coordinates if the temperature of the sample is low.

A fixed electric field F is now applied in the x direction; the carrier moving in the field enters a region in which its effective mass in the y direction be-

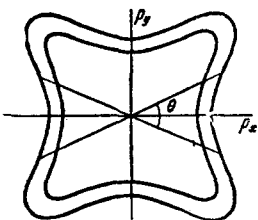
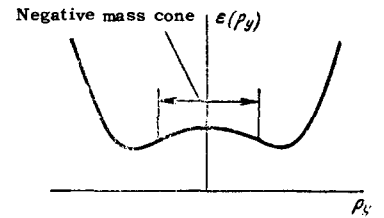


FIG. 10. Possible cross section for equipotential surfaces in a plane.

FIG. 11. Curve showing the dependence of energy ϵ on momentum p_y for $p_x = \text{const.}$



comes negative, so that it can amplify an oscillating electric field applied along the y axis. In this case negative-mass states are achieved at relatively small carrier energies so that losses due to ionization and emission of optical phonons are negligible.

It is proposed in reference 37, that under the effect of the field the carrier is displaced inside the negative mass cone, since scattering on the acoustic vibrations of the lattice is neglected; it is also assumed that upon reaching an energy equal to the energy of an optical phonon the carrier radiates instantaneously and returns to the origin of the coordinates. This assumption corresponds to an infinitely large optical-phonon interaction constant D^2 so that $D^2/C^2 \rightarrow \infty$. However, in actual semiconductors these constants are of the same order of magnitude and the distribution function is "spread out" in the direction of p_y . Although the distribution function has not been computed for this case, it would appear that it is again impossible to obtain a distribution which satisfies (5.4). Certain authors have proposed that radiating states with negative mass can be obtained by using fixed electric and magnetic fields simultaneously.⁴² It is shown in reference 41, however, that negative temperature states cannot be obtained in this way.

Thus, the application of fixed fields to a semiconductor cannot produce negative temperature states for transitions inside a single band. As in transitions between different bands, one might expect to produce negative temperatures within a single band by using a pulse technique in a semiconductor; however the practical realization of this idea is not very likely because times $t \approx 10^{-10} - 10^{-12}$ sec are involved.

6. Production of Negative Temperatures by Double-Resonance Methods

In maser amplifiers that operate at centimeter wavelengths negative-temperature states are frequently produced by allowing the quantum-mechanical system to interact with an auxiliary high-frequency (pumping) field.^{43,72,73}

Consider a three-level system (Fig. 12). Suppose that radiation of frequency ω_{13} is applied to the system and that the intensity of radiation is such that levels 1 and 3 are saturated; under these conditions we can produce a negative temperature between levels 1 and 2 or 2 and 3.

In reference 44 it was first proposed to obtain selection by means of an auxiliary high-frequency pump-

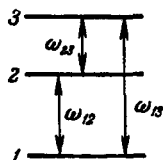


FIG. 12. Three-level system.

ing field in amplifiers in which spin systems are used.

In these systems the level populations are given by the equations

$$\left. \begin{aligned} \frac{dn_1}{dt} &= \omega_{13} \left(n_3 - n_1 + \frac{N\hbar\omega_{31}}{3kT} \right) + \omega_{21} \left(n_2 - n_1 + \frac{N\hbar\omega_{21}}{3kT} \right) \\ &\quad - W_{31} (n_1 - n_3), \\ \frac{dn_2}{dt} &= \omega_{23} \left(n_3 - n_2 + \frac{N\hbar\omega_{32}}{3kT} \right) + \omega_{21} \left(n_1 - n_2 - \frac{N\hbar\omega_{21}}{3kT} \right) \\ &\quad + W_{32} (n_3 - n_2), \\ \frac{dn_3}{dt} &= \omega_{13} \left(n_1 - n_3 - \frac{N\hbar\omega_{31}}{3kT} \right) + \omega_{23} \left(n_2 - n_3 - \frac{N\hbar\omega_{32}}{3kT} \right) \\ &\quad + W_{31} (n_1 - n_3) + W_{32} (n_2 - n_3), \end{aligned} \right\} \quad (6.1)$$

where n_i is the population of the i -th level, $N = n_1 + n_2 + n_3$, T is the temperature of the system, ω_{ij} is the frequency of the transition between the levels denoted by i and j and w_{ij} is the probability of a transition from level i to level j under the effect of thermal excitation. For thermal equilibrium

$$\frac{\omega_{ij}}{\omega_{ji}} = e^{-\frac{\hbar\omega_{ij}}{kT}}$$

for $i < j$ where W_{31} and W_{32} are the probabilities for the transition between levels 3-1 and 3-2 respectively. In Eq. (6.1) it is assumed that $\hbar\omega_{ij} \ll kT$.

When the intensity of the pumping radiation is high, so that $W_{31} \gg W_{32}$, an approximate solution of the system is (6.1) is

$$n_1 - n_2 = n_3 - n_2 = \frac{N\hbar(-\omega_{23}\omega_{32} + \omega_{21}\omega_{31})}{3kT(\omega_{23} + \omega_{31} + W_{32})} \quad (6.2)$$

The use of pumping radiation in the optical region for changing populations in Zeeman levels was first proposed in reference 45. If circularly polarized radiation is used, it is possible to effect a redistribution over the Zeeman levels since, transitions take place which satisfy the selection rule $\Delta s = +1$ or $\Delta s = -1$, depending on the direction of polarization (s is the quantum number that characterizes the projection of the angular momentum of the system in a given level in the field direction). For example, for the level system shown in Fig. 13, an excess population can be obtained in the $S_{1/2}$ level with $s = +\frac{1}{2}$ if circularly polarized radiation is used to induce transitions characterized by $\Delta s = +1$, provided the relaxation time between $s = \pm\frac{1}{2}$ levels is appreciably greater for $S_{1/2}$ than the lifetime of the atom in the excited state $P_{1/2}$, with $s = +\frac{1}{2}$; as a rule this condition is satisfied. In spontaneous emission the transitions $\Delta s = +1$ and $\Delta s = 0$ are equally probable, so that an excess popu-

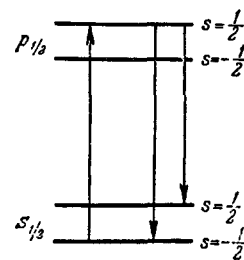


FIG. 13. Level diagram showing Zeeman splitting.

lation accumulates in the $S_{1/2}$, $s = +\frac{1}{2}$ level. In this case it is assumed that the relaxation time between the $P_{1/2}$, $s = \pm\frac{1}{2}$ is large compared with the lifetime in the $P_{1/2}$ level for transitions to the $S_{1/2}$ level. If it is small, a population accumulates in the $S_{1/2}$, $s = +\frac{1}{2}$ level, as before, since transitions from $P_{1/2}$, $s = -\frac{1}{2}$ to $S_{1/2}$, $s = \pm\frac{1}{2}$ are equally probable. The ratio of the number of atoms with $S_{1/2}$, $s = +\frac{1}{2}$ to the number of atoms with $S_{1/2}$, $s = -\frac{1}{2}$ is equal to the ratio of the relaxation times between these levels to the reciprocal probability for excitation from $S_{1/2}$, $s = -\frac{1}{2}$ to $P_{1/2}$, $s = +\frac{1}{2}$

$$\frac{n_2}{n_1} = \frac{1}{2} \tau W, \quad (6.3)$$

where n_2 is the number of atoms in the $S_{1/2}$, $s = +\frac{1}{2}$ state, n_1 is the number of atoms in the $S_{1/2}$, $s = -\frac{1}{2}$ state, W is the probability of excitation from $S_{1/2}$, $s = -\frac{1}{2}$ to $P_{1/2}$, $s = +\frac{1}{2}$ and τ is the lifetime of the atom in the $S_{1/2}$, $s = +\frac{1}{2}$ state.

The transition frequency for adjacent Zeeman levels is

$$\frac{\omega}{2\pi} = \frac{eH}{2\pi mc} \approx 2.9 \cdot 10^6 H \text{ sec}^{-1} \quad (6.4)$$

where H is the strength of the magnetic field in cgs units. In realizable fields $H \approx 10^5$ and $\omega/2\pi \approx 3 \times 10^{11} \text{ sec}^{-1}$, yielding radiation in the millimeter wavelength region. For this reason the Zeeman splitting cannot be used for obtaining radiation at infrared or optical wavelengths.

However, three-level quantum-mechanical systems of this kind can be used to transform optical and infrared radiation into radiation in the centimeter wavelength range, where high-sensitivity detectors are available.

It should be noted, that when semiconductors are considered, the quantity m in Eq. (6.4) is the effective mass of the carrier; the effective mass can be appreciably smaller than the mass of the electron. In this case the transition frequency ω in realizable fields of 10^5 oe can approach the limits of the infrared region.

A shortcoming of this method of obtaining negative temperatures is that presently available radiation sources are not powerful enough to achieve saturation in the pumping transition. For example, for a transition frequency $\omega/2\pi = 10^{14} \text{ sec}^{-1}$ ($\lambda \sim 3 \mu$), with a dipole moment $d \approx 10^{-18}$ cgs units and $\Delta\omega/\omega \sim 10^{-6}$, the power required for saturation is

$$P \approx \frac{cF^2}{4\pi} \approx \frac{(\Delta\omega)^2 ch^2}{4\pi d^2} \approx 10^5 \text{ w/cm}^2. \quad (6.5)$$

The pumping power can be reduced by several orders of magnitude if resonators are used; however, the required power is still quite high and in practice the pumping transition is far from saturation.

It follows from the system (6.1) that the ratio of the populations in levels 2 and 3 is independent of the pumping power; this ratio is given by

$$\frac{n_3}{n_2} = \frac{\omega_{21}}{\omega_{32}}. \quad (6.6)$$

This relation obtains when the population in levels 2 and 3 due to transitions caused by the pumping radiation is larger than the population characteristic of thermal equilibrium in these levels; that is to say, the minimum power required for producing a negative temperature is determined from the condition

$$\frac{\omega_{32} W n_1}{\omega_{31} (\omega_{32} + \omega_{31})} \gg n_2^0, \quad (6.7)$$

where W is the probability of a transition under the effect of the pumping radiation [cf. Eq. (6.1)] and n_2^0 is the population of level 2 at thermal equilibrium.

Using Eq. (7.3) for the transition probability, when the level width due to collisions is approximately equal to the Doppler width (in this case minimum pumping is required), we obtain the following inequality:

$$P > \frac{ch^2 \Delta\omega \omega_{21} (\omega_{32} + \omega_{31})}{2\pi |d_{13}|^2 \omega_{32}} e^{-\frac{h\omega_{13}}{kT}}. \quad (6.8)$$

For example, for the level scheme shown in Fig. 12, with $\omega_{13}/2\pi = 1 \times 10^{14} \text{ sec}^{-1}$, $\omega_{32}/2\pi = 3 \times 10^{13} \text{ sec}^{-1}$, $\Delta\omega/\omega \approx 10^{-6}$, $\omega_{31} = \omega_{32} = 10^7 \text{ sec}^{-1}$, $\omega_{21} = 10^8 \text{ sec}^{-1}$, $d_{13}^2 \approx 10^{-18} \text{ cgs esu}$, and $T \approx 300^\circ \text{K}$, the required power is

$$P > 10^{-3} \text{ w/cm}^2. \quad (6.9)$$

Thus, to obtain negative temperature states between levels 3 and 2 it is necessary, as follows from Eq. (6.6), that $\omega_{21} > \omega_{32}$; in addition, the pumping power must satisfy the condition in (6.8). When the transition frequency is increased, the required power is reduced, as follows from (6.8). The production of negative temperature states is thus facilitated; on the other hand the number of active molecules is reduced.

In reference 46 it is proposed that atomic levels in gaseous potassium be used to obtain a negative temperature in the wavelength range $\lambda \sim 3.14 \mu$. A level diagram for potassium, and the pertinent lifetimes, are shown in Fig. 14. In this case the pumping radiation causes transitions between the 4s and 5p levels while the negative temperature is maintained between the 5p and 3d levels. It is suggested that pumping radiation can be obtained from a potassium lamp by means of a filter which selects radiation corresponding to the 4s-5p transition while removing radiation corresponding to the 4s-4p transition. The vapor pressure of the

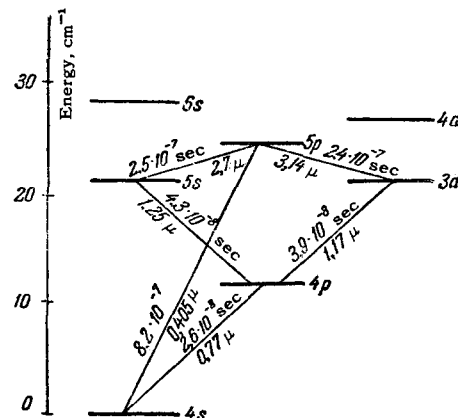


FIG. 14. Level diagram for potassium.

potassium vapor must be such that the line width is due primarily to the Doppler effect. The required potassium vapor pressure can be achieved at $T = 435^\circ \text{K}$ and in this case $\Delta\omega/\omega = 1.2 \times 10^{-6}$. In reference 46 it is stated that sufficient pumping power can be obtained with a 15-watt potassium lamp operated with forced cooling. According to the authors of reference 46, the radiation power at the required frequency (4s-5p transition) is 0.6 mw for a lamp volume of 5 cm^3 .

In reference 47 the use of pumping radiation is considered for obtaining negative temperature states in solids; in solids the level width is much broader than in gases and for this reason the power required to obtain negative temperature is greater than that given in (6.5).

If the pumping power source has a broad spectrum, and the lines are broad, as is the case in solids, somewhat less power can be used. Nevertheless the power of presently available sources is still inadequate for obtaining negative temperatures in solids.

The most promising materials for obtaining negative temperatures in solids would seem to be single-crystals that contain impurity atoms of the rare earths; these have narrow spectral lines associated with electron transitions between levels in unfilled inner shells.

It is interesting to note that recently obtained spectra of certain organic materials at liquid-nitrogen temperatures exhibit lines with widths as narrow as 1 or 2 cm^{-1} .^{48a} These materials may turn out to be suitable for obtaining negative temperature states by the double-resonance method.

An experimental investigation of the possibility of obtaining a negative temperature state between the 8^2D and 7^2P levels in cesium is reported in reference 79.

The cesium atoms are excited from the $6^2\text{S}_{1/2}$ level to the $8^2\text{P}_{1/2}$ level by a helium lamp. If inert gases (for example, helium) are added to the cesium vapor, the cesium atoms make transitions from the $8^2\text{P}_{1/2}$ level to the $8^2\text{D}_{3/2}$ level; it then becomes possible to produce a negative temperature between the 8^2D and 7^2P levels.

In the experiments the authors observed that the intensity of the radiation from the cesium atoms was increased when the intensity of the excitation radiation from the helium lamp was increased. This effect can be explained by a negative temperature between the 8^2D and 7^2P levels in cesium. However, the authors point out that the experiment does not give really direct verification of a negative temperature in the cesium.

II. GENERATION AND AMPLIFICATION OF ELECTROMAGNETIC WAVES BY NEGATIVE TEMPERATURE SYSTEMS

7. Interaction of Radiation with Negative-Temperature Systems

A system in a negative-temperature state amplifies radiation incident upon the system. If certain conditions are satisfied — the oscillation conditions, the negative-temperature system becomes unstable and oscillates.¹

By analogy with positive-temperature systems, which are characterized by an absorption coefficient, a system in a negative-temperature state can be characterized by an emission coefficient $\alpha(\omega)$ which describes the growth of the amplitude squared (radiation intensity) $I = cF^2/4\pi$ of the wave transmitted through the system. This problem has been considered to some extent in reference 79. For small values of intensity, in which case saturation can be neglected, the following relation holds:

$$I(l) = I(0) e^{+\alpha(\omega, I)l}, \quad (7.1)$$

where $I(0)$ is the initial value of the field intensity, at the point $x = 0$, and $I(l)$ is the same quantity at $x = l$, where l is the length of path traversed by the wave. The emission coefficient¹² $\alpha(\omega)$ is related to the probability of a stimulated transition W_{mn} by the following expression:

$$\alpha(\omega, I) = \frac{W_{mn} \hbar \omega n}{I}, \quad (7.2)$$

$$W_{mn} = \frac{1/\tau}{(\omega - \omega_{mn})^2 + 1/\tau^2 + d_{mn}^2 F^2 / \hbar^2} \frac{d_{mn}^2 F^2}{2\hbar^2}, \quad (7.3)$$

where ω_{mn} is the frequency of the transition, ω is the frequency of the incident radiation, d_{mn} is the matrix element of the dipole moment between the levels being considered, τ is the mean lifetime of the system in the excited state, and n is the number of active molecules per unit volume.*

At high intensities (saturation), the inequality

$$\frac{1}{\tau^2} \ll \frac{d_{mn}^2 F^2}{\hbar^2}, \quad (7.4)$$

holds in (7.3), and the dependence of intensity on distance is more complicated. In the limiting case of

*Equation (7.3) applies when the line broadening associated with the lifetime τ is greater than that due to other causes, for example, the Doppler effect in beams, wide carrier bands in semiconductors, etc.

strong saturation

$$I(l) = I(0) + Al. \quad (7.5)$$

The spectral distribution of radiation which passes through such a medium is also affected. Because the direction, polarization, and frequency of the photons produced in stimulated emission are exactly the same as those of the incident photons, any change in the spectral distribution of the radiation is due to the line shape alone.

Suppose that broadband low-intensity radiation is incident on a system, so that (7.1) holds.

At distances $l < 1/\alpha(\omega_{mn})$, we add to the initial intensity $I(\omega, 0)$ an intensity with a spectral composition in the form of the spectral line

$$I(\omega, l) = I(\omega, 0) [1 + \alpha(\omega)l]. \quad (7.6)$$

At distances $l \gg 1/\alpha(\omega_{mn})$, before saturation sets in, the spectral distribution of the radiation is much narrower than the spectral line (Fig. 15). The width is given by

$$\frac{1}{\tau'} = \frac{1}{\tau} \sqrt{\frac{\ln 2}{\alpha(\omega_{mn})l}}. \quad (7.7)$$

At high intensities (saturation), the spectral distribution of the radiation stimulated by the incident photons is retained (Fig. 15).

Thus, when spontaneous emission is produced in a sample of sufficiently large dimension, and the spectral distribution of the spontaneous emission is given by the line, the output from the sample is essentially monochromatic at a frequency close to the peak of the spectral line associated with the spontaneous emission.

In the analysis given above we neglect reflection of the radiation at the boundaries of the sample, i.e., it is assumed that there is no feedback in the system. In a certain sense the radiation produced under these conditions is similar to fluorescence radiation; it is distinguished from the latter, however, by the narrower spectral distribution.

In addition to the process described above, in which the number of photons is increased by stimulated emission, the sample exhibits radiation due to spontaneous transitions. The overall spectral distribution of the radiation (spontaneous and stimulated) can be obtained as follows (Fig. 16).

We consider the radiation produced by spontaneous and induced transitions in a plane layer dx ; for the assumed nonsaturation conditions

$$\frac{dI(\omega)}{dx} = \alpha(\omega)I(\omega) + \beta(\omega), \quad (7.8)$$

where $\beta(\omega)$ is the intensity of the spontaneous emission per unit length of sample in the x direction in the solid angle of interest to us do. Integrating, we have

$$I(\omega) = \tilde{I}(\omega) e^{\alpha l} - \frac{\beta(\omega)}{\alpha(\omega)}, \quad (7.9)$$

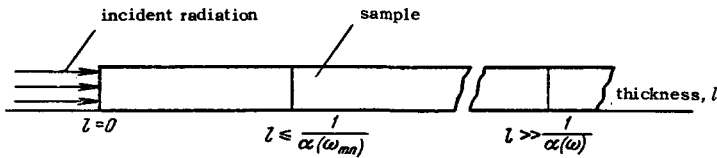


FIG. 15. Narrowing of the spectral distribution of radiation transmitted through a medium with negative losses in the absence of saturation.

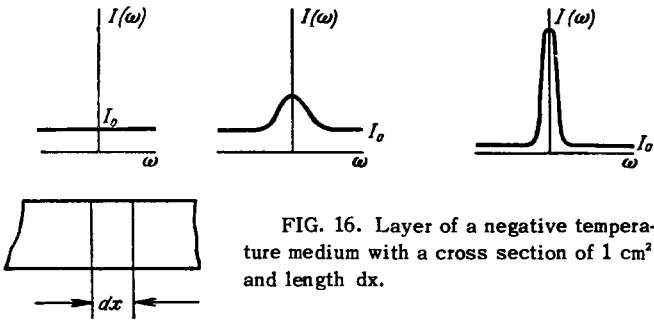


FIG. 16. Layer of a negative temperature medium with a cross section of 1 cm^2 and length dx .

where

$$\tilde{I}(\omega) = I(\omega, 0) + \frac{\beta}{\alpha}.$$

Thus, for a thick sample $l \gg 1/\alpha(\omega_{mn})$ the spectral distribution consists of a very narrow line $\tilde{I}(\omega)e^{\alpha l}$ superimposed on the background due to spontaneous emission $\beta(\omega)/\alpha(\omega)$.

It should be noted that while the width of the stimulated emission line is reduced as the sample dimensions are increased, the spectral distribution of the spontaneous emission background is independent of sample dimensions.

For saturation conditions, Eq. (7.8) is replaced by the following expression:

$$\frac{dI(\omega)}{dx} = C + \beta, \quad (7.10)$$

where

$$C = \frac{n_0 \hbar \omega}{2\tau} \quad \text{for} \quad (\omega - \omega_{mn})^2 < \frac{d_{mn}^2 F^2}{\hbar^2}.$$

Integrating Eq. (7.10) we have

$$I(\omega, l) = I(\omega, 0) + (C + \beta)l. \quad (7.11)$$

In this case the narrow spectral line $I(\omega, 0)$ is superimposed on the broad background, whose intensity increases linearly with sample dimensions.

As the thickness of the sample is increased the radiation approaches the continuous background while the intensity of the spectral line (superimposed on this background) is reduced.

It should be pointed out that for sufficiently large sample dimensions the spectral distribution of the thermal radiation due to a spectral line of width $\Delta\omega$ will be considerably broader than $\Delta\omega$.

8. Oscillation Condition

In the preceding section we have considered the transmission of radiation through a negative-temperature medium without taking account of effects which

arise at the boundaries of the sample. When these effects are neglected the radiation is similar to fluorescence. If there are reflecting boundaries in a negative temperature system, however, there is a positive feedback effect: photons reflected from the boundaries can return to the point in space from which they originate in the same state in which they were emitted from this point. Because of this positive feedback, the system can oscillate when certain conditions (oscillation condition) are satisfied.

For a maser oscillator containing a resonator with quality factor Q_{res} in which one oscillation mode can be excited, the oscillation condition¹ is written

$$\beta = \frac{4\pi N_0}{V\hbar} |d|^2 Q_{\text{res}} \tau^2 > 1, \quad (8.1)$$

where β is the feedback factor (for oscillation $\beta > 1$), N_0 is the number of active molecules which enter the resonator per second, V is the volume of the resonator, $|d|$ is the dipole moment matrix element and $1/\tau = \Delta\omega$ is the width of the spectral line. It is convenient to rewrite Eq. (8.1) in the symmetric form

$$\beta = \frac{4\pi n_0 |d|^2}{\hbar\omega} Q_{\text{res}} Q_{\text{line}} > 1, \quad (8.2)$$

where $n_0 = N_0\tau/V$ is the number of active molecules per unit volume

$$Q_{\text{line}} = \frac{\omega}{\Delta\omega}.$$

In the optical and infrared regions it is essentially impossible to excite only one mode in the neighborhood of a given frequency, because the linear dimensions of a typical resonator are of the order of the radiation wavelength. Hence, in the optical and infrared wavelength regions we must consider resonators in which higher modes are excited simultaneously, so that a given frequency corresponds to several modes. Under these conditions, just as in the one-mode case, self-excitation or oscillation is achieved when the power radiated in a given mode by the medium with negative losses is greater than the energy loss in this mode, i.e., when

$$P_{\text{rad}} > P_{\text{loss}}, \quad (8.3)$$

where P_{rad} is the power radiated by the negative

temperature system and P_{loss} is the power lost in the resonator.

The power lost in the resonator can be expressed in terms of the quality factor for a given mode

$$P_{\text{loss}} = \frac{E_{\text{stor}}\omega}{Q_{\text{res}}},$$

where $E_{\text{stor}} = \hbar\omega N$ is the energy stored in the resonator and N is the number of photons in the system for a given mode.

The power radiated by the system can be computed from the spontaneous decay lifetime. Let the lifetime for radiative decay of a system of p oscillators in a frequency band given by the line width $\Delta\omega$ be τ ; the probability of radiative decay in a given mode is then

$$W_c = \frac{1}{p\tau}. \quad (8.4)$$

The probability of stimulated emission W_s under the effect of N photons in a given mode is N times greater than the probability given in (8.4):

$$W_s = \frac{N}{p\tau}. \quad (8.5)$$

If a system of volume V contains nV active molecules, the power radiated in a given mode is

$$P_{\text{rad}} = \frac{\hbar\omega nV}{p\tau}. \quad (8.6)$$

Thus, the oscillation condition (8.3) leads to the expression

$$\frac{\hbar\omega NnV}{p\tau} > \frac{\omega}{Q_{\text{res}}} N\hbar\omega, \quad (8.7)$$

i.e.,

$$\beta = \frac{Q_{\text{res}} nV}{\omega p\tau} > 1.$$

If the volume V is free space, the quantity p is

$$p = \frac{8\pi\omega^2\Delta\omega V}{(2\pi)^3 c^3}. \quad (8.8)$$

In semiconductor oscillators the lifetime τ depends on the number of free carriers n and can be expressed in terms of τ_0 the value for some carrier concentration n_0 :

$$\tau = \frac{n_0\tau_0}{n}. \quad (8.9)$$

The condition in (8.7) now assumes the form

$$\frac{Q_{\text{res}} n^2 V}{\omega p n_0 \tau_0} > 1. \quad (8.10)$$

As we have noted in the introduction, the dimensionless quantity that determines the quality of a negative temperature system is

$$\kappa = |d|^2 \frac{2\pi n}{\hbar\Delta\omega}, \quad (8.11)$$

where $|d|^2$ is the square of the dipole-moment matrix element, n is the difference in the populations in the upper and lower levels, and $\Delta\omega$ is the width of the spectral line.

The square of the dipole moment matrix element is related to the spontaneous decay lifetime by the expression

$$\tau = \frac{3\hbar c^3 (2\pi)^3}{32\pi^3 \omega^3 |d|^2}. \quad (8.12)$$

In gas systems $\Delta\omega/2\pi \approx 2 \times 10^7$ cps at a pressure of the order of 1 mm Hg, $|d|^2 \approx 10^{-36}$ cgs esu, and the maximum number of active molecules for a given rotational level can equal the total number of molecules in the rotational level, i.e., $n = n_1/\Sigma_{\text{rot}}$ where n_1 is the total number of molecules per cubic centimeter at a pressure of 1 mm Hg and Σ_{rot} is the rotational partition function. For $\Sigma_{\text{rot}} \approx 10^3$ in a gas we find $\kappa_{\text{gas}} \leq 10^{-3}$. For paramagnetic ions in a crystal $\Delta\omega \sim 10^{-2}\omega$, $\omega/2\pi \sim 10^{11}$, $|d|^2 \approx 10^{-38}$ and for a paramagnetic ion concentration of approximately 1% we find $\kappa_p \leq 0.1$.

When considering semiconductor materials, it is convenient to replace $|d|^2$ in Eq. (8.11) by the lifetime for radiative transitions (8.12). Substituting τ from Eq. (8.9) in Eq. (8.12) we obtain the following expression for κ :

$$\kappa = \frac{3\pi c^3 n^2}{2\pi^3 \omega^3 \tau_0 n_0 \Delta\omega}. \quad (8.13)$$

In this case κ is a nonlinear function of the density of active particles n .

For the semiconductor crystal InSb, in which $\frac{\Delta\omega}{\omega} \approx 10^{-1}$, $\frac{\omega}{2\pi} \sim 3 \cdot 10^{13}$ sec $^{-1}$, $n \sim 10^{17}$, $\tau_0 \approx 10^{-6}$ sec

for $n_0 = 5 \cdot 10^{16}$
 κ is²¹

$$\kappa \approx 1.$$

The power P radiated by the maser oscillator when the oscillation condition is well satisfied is determined by the number N_0 of active molecules produced per unit time:¹

$$P_{\text{max}} = \frac{N_0}{2} \hbar\omega. \quad (8.14)$$

When a semiconductor system is pulsed, the maximum power is

$$P'_{\text{max}} = \frac{n}{2} \frac{\hbar\omega}{\Delta t}, \quad (8.15)$$

where Δt is the length of the radiation pulse (the order of the electron thermalization time in the semiconductor if the thermalization time is shorter than the lifetime).

In cw maser oscillators the bandwidth is of the order of the frequency spread $\delta\omega$ due to spontaneous emission:⁴⁹

$$\delta\omega = \frac{2kT(\Delta\omega_{\text{sp}})^2}{P}. \quad (8.16)$$

In pulse operation the bandwidth is determined by the radiation pulse Δt since $\delta\omega \ll 1/\Delta t$.

In the infrared and optical regions, radiation sources are conventionally characterized by an effective temper-

ature. The effective temperature of an oscillator can be obtained if we assume that the power radiated by the oscillator in the frequency interval determined by the bandwidth (8.16) is equal to the power radiated in the same frequency interval by a black body. The energy flux from a square centimeter of surface of a black body at a temperature T is given by the Rayleigh-Jeans law when $kT \gg \hbar\omega$

$$S = \frac{kT\omega^2\Delta\omega}{(2\pi c)^2}. \quad (8.17)$$

For a semiconductor cube with sides l

$$T_{\text{eff}} \approx \frac{2\pi n\hbar c^2}{k\omega}. \quad (8.18)$$

An estimate of the effective temperature for the case $n = 10^{17} \text{ cm}^{-3}$, $l \approx 1 \text{ cm}$, and $\omega/2\pi \approx 3 \times 10^{13}$ yields

$$T_{\text{eff}} > 10^{13} \text{ degrees.}$$

In the present section we have considered the oscillation condition when the resonator can support several modes characterized by quality factors which are approximately the same. The case in which oscillation takes place in several different modes simultaneously requires separate analysis.⁵⁰

9. Resonators

As has been noted above, simple modes cannot be used in a resonator in the infrared and optical regions because the resonator dimensions would be small and the quality factor would be low for this kind of operation.

A resonator consisting of two parallel reflecting plates has been suggested in reference 13. The quality factor for this configuration has been computed and found to be rather high. This kind of resonator is analogous to the Fabry-Perot interferometer used in optical work. An experimental investigation of this resonator in the millimeter wavelength region is reported in reference 76.

A more detailed analysis of resonators of this kind, which takes the spontaneous noise characteristic into account, is given in reference 46. If the radiation wavelength is small compared to the distance between the plates and the dimensions of the plates, the geometric-optics approximation can be used. In order to compute the quality factor of a resonator of this kind we must estimate the energy losses caused by reflection at the walls and by radiation through the lateral surfaces. The energy lost by reflection at the walls is

$$dE = E \frac{cdt}{L} (1 - \alpha), \quad (9.1)$$

where E is the total energy in the resonator; cdt/L is the number of reflections from the walls (separated by distance L) in a time dt ; α is the reflection coefficient; c is the velocity of light. Integrating Eq. (9.1) we have

$$E = E_0 e^{-\frac{c(1-\alpha)t}{L}} \quad (9.2)$$

Consequently the characteristic damping time (photon lifetime) is

$$t_0 = \frac{L}{c(1-\alpha)}. \quad (9.3)$$

This lifetime is related to the resonator bandwidth:

$$\Delta\omega = \frac{1}{t_0}, \quad (9.4)$$

which can be expressed in terms of the quality factor Q of the resonator

$$\Delta\omega = \frac{\omega}{Q}. \quad (9.5)$$

Thus,

$$Q = t_0\omega = \frac{L\omega}{c(1-\alpha)}. \quad (9.6)$$

An estimate of the quality factor for $L = 1 \text{ cm}$, $\alpha = 0.95$, and $\omega/2\pi = 10^{14} \text{ sec}^{-1}$ ($\lambda = 3 \mu$) yields

$$Q \approx 4 \cdot 10^5.$$

In addition to the energy losses due to reflection from the plates, there is a loss of energy due to radiation through the lateral surfaces of the resonator (Fig. 17)*

$$dE = -E \frac{c \tan\theta dt}{D}, \quad (9.7)$$

where D is the transverse dimension of the resonator and θ is the diffraction angle.

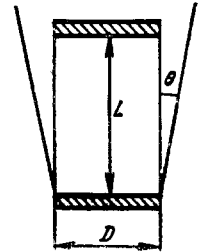


FIG. 17. Energy loss due to radiation through the lateral surfaces of a resonator.

From Eq. (9.7) it follows that ($2\theta \approx \lambda/D \ll 1$)

$$E = E_0 e^{-\frac{c\theta t}{D}}, \quad (9.8)$$

i.e.,

$$t'_0 = \frac{D}{c\theta} = \frac{2D^2}{c\lambda}. \quad (9.9)$$

Consequently

$$Q' = \frac{D^2\omega^2}{\pi c^2}. \quad (9.10)$$

An estimate of the value of Q' for $D = 1 \text{ cm}$ and $\omega/2\pi = 10^{14} \text{ sec}^{-1}$ yields $Q \approx 10^8$. It is obvious from this value that diffraction is not important for the conditions described here.

In resonators of this type, high Q can be obtained only when the parallelism of the plates is very good. In practice, the Q is determined by the parallelism of the plates.

*To reduce the loss and the number of resonator modes, the lateral walls can be made of a dielectric material; this approach has been suggested in reference 78.

If the plates are not strictly parallel, the deviation angle is increased by an amount β_0 for each reflection (Fig. 18). Thus, the beam "walk-off" time due to non-parallelism is of order

$$t_0'' = \eta \frac{L}{c}, \tag{9.11}$$

where η is the maximum number of reflections the

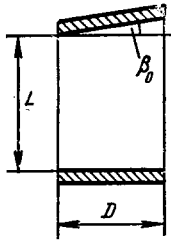


FIG. 18. Energy loss due to nonparallelism of the plates.

beam can experience before leaving the system; this quantity is determined by the condition

$$\sum_1^\eta \beta_0 n L = D. \tag{9.12}$$

Then

$$t_0'' = \frac{1}{c} \left(\frac{2LD}{\beta_0} \right)^{\frac{1}{2}}. \tag{9.13}$$

The quality factor is

$$Q'' = \frac{\omega}{c} \left(\frac{2LD}{\beta_0} \right)^{\frac{1}{2}}. \tag{9.14}$$

For $Q'' \approx 10^6$, $L = D = 1$ cm, and $\omega/2\pi = 10^{14}$ sec⁻¹ we have $\beta_0 \approx 10^{-3} \approx 3''$. The high resonator Q obtained at short wavelengths shows that it is completely feasible to use dielectric resonators; in these resonators reflection occurs at the boundary between two media with different dielectric constants. In a semiconductor oscillator the resonator can be the material itself, since semiconductors have high dielectric constants ($\epsilon \sim 10$) up to the infrared region. On the other hand, the high dielectric constant of semiconductors means that it is difficult to extract the radiation from the material, because the radiation incident on the surface at oblique angles experiences total internal reflection. Hence, modes which propagate at small angles to the surface of the sample have high Q and it is reasonable to assume that oscillation will take place primarily in these modes. The problem of extracting radiation from systems which support a large number of modes is one of considerable difficulty.

To compute the noise in maser amplifiers we must know the number of modes which have high Q at a given frequency. We have shown above that the Q associated with reflection from the resonator walls is smaller than that due to "divergence" resulting from diffraction. Because of this relation between the quality factors, a rather large number of modes which propagate within some angle ϑ , which is greater than the diffraction angle, will have high Q.

By analogy with Eq. (9.9) we can write the lifetime for photons reflected from the resonator surface at an angle ϑ

$$t_0 = \frac{L}{c\vartheta}. \tag{9.15}$$

We equate Eq. (9.15), which gives the reflection lifetime, and Eq. (9.3), to find the angle ϑ , inside of which the quality factor is large,

$$\vartheta = \frac{(1-\alpha)D}{L} = \frac{\omega D}{Qc}. \tag{9.16}$$

It follows that the number of high-Q modes in a resonator is equal to the total number of modes in a volume LD^2 , multiplied by the solid-angle ratio ($2\pi\vartheta^2/4\pi$)

$$P_{res} = p \frac{\vartheta^2}{2}. \tag{9.17}$$

From Eqs. (9.8), (9.16), and (9.17) we have

$$P_{res} = \frac{LD^4\omega^4\Delta\omega}{2\pi^2c^5Q^2}. \tag{9.18}$$

For the kind of resonator considered above ($L = D = 1$ cm, $\Delta\omega/\omega = 10^{-3}$, and $\omega/2\pi = 10^{14}$ sec⁻¹), we find

$$P_{res} \approx 10^6. \tag{9.19}$$

It is desirable to reduce the surface of the reflecting plates in order to reduce the number of modes; for example, when $D = 0.1$ cm the number of modes is reduced to 10^2 .

Coupling to a single mode, or a small number of modes, can be achieved by means of the diffraction pattern at the edge of the plate⁴⁶ and a lens (Fig. 19).

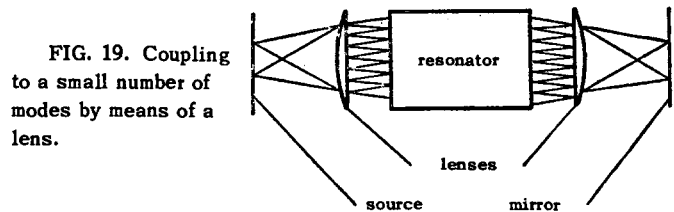


FIG. 19. Coupling to a small number of modes by means of a lens.

Since parallel bundles of rays incident on the lens at different angles are focused at different points in the focal plane, a small-aperture mirror located in the focal plane can be used to obtain a coupling with single modes. Below we consider the conditions that must be satisfied to obtain coupling to one mode by means of a lens. Suppose the radiation from the resonator passes through one of the plates of the resonator. If the reflection coefficient is α , the extracted power is smaller than the power incident on the wall by a factor $(1 - \alpha)$.

For mode separation to be realized, the angle between the propagation directions of neighboring modes must be larger than some minimum value. This angle can be estimated as follows. A standing wave exists between the two resonator plates, separated by a distance L , when the length L is a multiple of an integral

number of half-wavelengths, i.e.,

$$2L = q\lambda, \quad (9.20)$$

where q is an integer.

For radiation propagated at a small angle to the normal to the surface of the plate, the analogous condition can be written in the form

$$\frac{2L}{\cos \theta_1} = q_1 \lambda. \quad (9.21)$$

It is obvious that the minimum angle θ_1 in Eq. (9.2) corresponds to the value $q_1 = q + 1$. This angle is precisely equal to the angle between the propagation directions of neighboring modes

$$\theta_1 = \left(\frac{\lambda}{L} \right)^{\frac{1}{2}}. \quad (9.22)$$

If the resonator has a finite Q , and thus a finite transmission bandwidth, a given mode does not propagate at a given angle, but rather within a range of angles $\Delta\theta_1$, which can be obtained in similar fashion. From the condition

$$\frac{2L}{\cos \Delta\theta_1} = q(\lambda - \Delta\lambda), \quad (9.23)$$

$$\Delta\lambda = -2\pi c \frac{\Delta\omega}{\omega^2} \quad (9.24)$$

we can show that

$$\Delta\theta_1 = \sqrt{\frac{2}{Q}},$$

where Q is the quality factor of the resonator; the quality factor and maximum bandwidth $\Delta\omega$ are related by the expression

$$Q = \frac{\omega}{\Delta\omega}. \quad (9.25)$$

If neighboring modes are to be separated the condition $\theta_1 > \Delta\theta_1$ must be satisfied; this condition means that

$$4\pi(1 - a) < 1. \quad (9.26)$$

In the derivation of Eq. (9.26) it is assumed that the quality factor of the resonator is determined by the reflection losses [Eq. (9.6)].

Besides (9.26), another condition must be satisfied if different modes are to be separated by means of a lens: the radiation which passes through the face plate of the resonator suffers diffraction because of the finite dimensions of this plate D , and the diffraction angle must be smaller than the angle between neighboring modes θ_1 . This requirement yields the condition

$$\frac{D^2}{\lambda} \gg L, \quad (9.27)$$

which is easily satisfied even when $D < L$, because in practice $D \gg \lambda$ in the infrared region. The requirements in (9.26) and (9.27) are easily satisfied in the resonators for which the numerical estimates have been carried out above; that is to say, it is possible to couple to a single mode.

10. Amplifiers

As indicated above, quantum-mechanical systems in negative-temperature states can be used to amplify electromagnetic waves.

Several kinds of amplifiers (paramagnetic amplifiers) have already been developed for the centimeter and decimeter ranges.⁵¹⁻⁵⁹ The distinguishing feature of these amplifiers is the extremely high sensitivity. The high sensitivity is a result of the fact that these amplifiers can operate at temperatures close to absolute zero, so that the amplifier noise is due only to spontaneous emission from excited ions.⁶⁰⁻⁶⁴

In principle, in the infrared wavelength region, just as in the centimeter region, two kinds of amplifiers can be used: 1) resonance amplifiers⁵¹ and 2) traveling-wave amplifiers.^{52,56,58}

A resonance amplifier is essentially a regenerative amplifier in which feedback is realized through the radiation field. The negative-temperature medium plays the role of the amplifier. The power gain of an amplifier of this kind is given by the feedback factor [cf. Eq. (8.1)]

$$\beta = \frac{4\pi N_0}{V\hbar} |d|^2 Q_{\text{res}} \tau^2.$$

The gain is⁶⁵

$$k = \frac{1}{(1-\beta)^2} k_0, \quad (10.1)$$

where k_0 is the power transfer function of the resonator in the absence of the negative temperature medium. The bandwidth of these amplifiers is reduced as the gain increases, as in other regenerative amplifiers. The nominal bandwidth of the amplifier is determined either by the width of the spectral line or by the resonator bandwidth, depending upon which is narrower. The dependence of bandwidth on gain is given by

$$\Delta\omega(k) = \sqrt{\frac{k_0}{k}} \frac{1}{\tau}. \quad (10.2)$$

The expressions given above apply for a resonator in which one mode is excited in the neighborhood of the frequency being considered.

As has been noted above, it is impossible in practice, at infrared wavelengths to make a resonator in which only one mode is excited at a given frequency. On the other hand, because the Q is high in the infrared region, several modes must be used if adequate bandwidth is to be obtained.

Let us assume that by means of some optical system we couple to M different modes (Fig. 20) that have approximately the same quality factors and lie within a frequency interval $\Delta\Omega$.

If the quality factors of the individual modes are such that these modes overlap in frequency, as shown in the figure, the transmission bandwidth remains unchanged as the gain factor k is varied so long as the

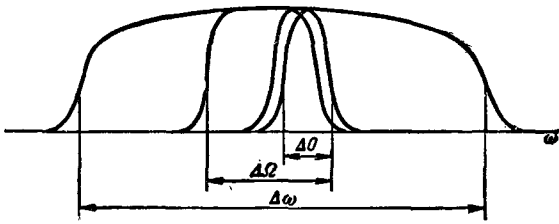


FIG. 20. Bandwidth of a multimode amplifier: $\Delta\Omega$ is the frequency interval over which the optical system is coupled, ΔO is the bandwidth of the resonator for a given mode, $\Delta\omega$ is the frequency spread of the radiative transitions in the medium in a negative temperature state.

modes do not separate; when separation occurs the gain exhibits dips at certain frequencies.

In essence, the simplest type of travelling wave amplifier has been considered in Sec. 7. As was indicated there, the gain and the bandwidth of this amplifier are determined by the width of the spectral line and by the radiation coefficient, and depend on the optical path traversed by the wave. The dimensions of amplifiers in the centimeter wavelength range are reduced by means of so-called slow-wave structures, in which the group velocity of the wave is small.⁵⁸ Systems of this kind can also be used in the infrared region (Fig. 21).

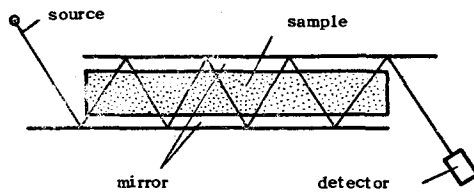


FIG. 21. Slow-wave system.

The noise in negative-temperature amplifiers can be determined, as noted above, by the spontaneous emission of the excited particles, if the temperature of the remaining elements of the amplifier is low enough. Below we estimate amplifier noise temperature.

For simplicity, we shall consider a two-level system with all the elements in the upper energy level. The noise is a maximum in this case.*

Suppose that the system is located in a resonator with quality factor Q and that one mode is excited at the frequency considered. We also assume that the width of the radiation line of the negative temperature system is greater than the resonator bandwidth. The radiation produced in the resonator is measured by a detector which does not introduce additional noise (for example, a noisy detector coupled to a resonator by an independent element with a ferrite isolator which operates at low temperatures). If the oscillation condition is not satisfied and the system operates as an

*This applies for noise due to coherent spontaneous emission, the possibility of which has been indicated in reference 66, cf. reference 77.

amplifier with gain k , the noise temperature can be determined as follows. The radiation power measured by the detector is increased if the temperature of the resonator walls is increased because at nonzero resonator temperatures there are thermal photons in the resonator and these are amplified by the amplifier. In accordance with the usual convention, the noise temperature is defined as that resonator temperature for which the noise power measured at the detector is doubled.

For a given resonator temperature T , the number of photons in a given mode is

$$n = (\frac{\hbar\omega}{e^{kT}} - 1)^{-1}. \quad (10.3)$$

The probability of stimulated emission due to the thermal photons becomes equal to the probability for spontaneous emission when $n = 1$; this relation then determines the noise temperature in question:

$$T_{\text{noise}} = \frac{\hbar\omega}{k \ln 2}. \quad (10.4)$$

The sensitivity of the amplifier can also be characterized by the number of photons which produce an output signal twice as large as the amplifier noise. In this case enough power must be introduced into the resonator to produce a single photon. By definition the quality factor is

$$Q = \frac{\omega E_{\text{stor}}}{P_{\text{loss}}}, \quad (10.5)$$

where E_{stor} is the energy stored in the resonator, P_{loss} is the power lost in the resonator, ω is the frequency of a given mode in the resonator. Taking $E_{\text{stor}} = \hbar\omega$ and $P_{\text{loss}} = \eta\hbar\omega$ (η is the number of photons produced in the resonator per second), we find

$$\eta = \frac{\omega}{Q}. \quad (10.6)$$

When the resonator supports several modes Eq. (10.6) becomes

$$\eta = \frac{M\omega}{Q}, \quad (10.7)$$

where M is the number of modes at frequency ω . The factor M in Eq. (10.7) arises because when several modes can be excited the spontaneous emission in each mode is independent of that in other modes.

It should be noted that the noise temperature, in the sense defined by Eq. (10.4), does not depend on the number of modes in the resonator; on the other hand the quantum sensitivity (10.7) is reduced as the number of modes increases.

Actually, in detection of the thermal radiation from a hot body (for example, the walls of the resonator itself), the number of photons in the resonator increases in proportion to the number of resonator modes. However, spontaneous emission noise, which is proportional to the number of different modes, also increases. Consequently, the sensitivity of a quantum mechanical amplifier with a multimode resonator (in which all modes are excited) does not change as the number of modes

is changed. However, since the number of photons in the resonator increases in proportion to the number of modes, the quantum sensitivity of the amplifier is reduced. For this reason, it is desirable to increase sensitivity by using single-mode amplifiers when detecting coherent external radiation.

The reduction in quantum sensitivity as the number of amplifier modes is increased can be characterized by an effective noise temperature; according to Eqs. (10.4) and (10.7) this temperature is

$$T_{\text{eff}} = T_{\text{noise}} M. \quad (10.8)$$

When the width of the spectral line is smaller than the resonator bandwidth, the noise temperature of the amplifier is reduced by a factor Q_{line}/Q .⁶⁷ This reduction in noise arises as follows: when the width of the spectral line is greater than the resonator bandwidth, all the photons are produced by stimulated emission of molecules; on the other hand when the line width is smaller, stimulated emission is responsible for only those photons which have frequencies within the width of the spectral line. In this case the noise temperature is

$$T'_{\text{noise}} = \frac{\hbar\omega}{k} \frac{\Delta\omega_{\text{line}}}{\Delta\omega_{\text{res}}}, \quad (10.9)$$

where $\Delta\omega_{\text{line}}$ is the width of the spectral line and $\Delta\omega_{\text{res}}$ the bandwidth of the resonator. Equation (10.9) shows that the noise temperature associated with spontaneous emission is reduced as the quality factor of the resonator is reduced; in the limiting case, $\Delta\omega_{\text{res}}/\omega \approx 1$, Eq. (10.8) becomes

$$T_{\text{noise}} = \frac{\hbar\Delta\omega_{\text{line}}}{k}, \quad (10.10)$$

that is to say, T_{noise} is determined by the bandwidth of the amplifier, as in conventional amplifiers. Because resonator quality factors are rather high in the infrared, and the quality factors of the spectral lines are low, it is difficult to realize the conditions described by Eq. (10.10).

Thus, noise temperature increases with increasing frequency; at $\omega/2\pi \approx 3 \times 10^{13} \text{ sec}^{-1}$ we find $T_{\text{noise}} \approx 1500^\circ$ for an amplifier with single-mode coupling. In multimode coupling the sensitivity of the amplifier is inversely proportional to the number of modes.

For the noise temperature given above $T_{\text{noise}} \approx 1500^\circ$, the power sensitivity of the quantum-mechanical amplifier is (assuming one mode)

$$P_{\text{line}} = kT_{\text{noise}} \frac{\Delta\omega}{2\pi} \approx 6 \cdot 10^{-12} \text{ w}$$

for a bandwidth of $\Delta\omega \approx 2 \times 10^9 \text{ cps}$; this sensitivity is considerably greater than that of bolometers usually used in the infrared region,⁶⁸ which have sensitivities of the order of 10^{-11} w and bandwidths of $\Delta\omega/\omega \sim 10^{-3}$. In this wavelength range the sensitivity of quantum-mechanical amplifiers is a thousand times greater than that of bolometers.

11. Quantum-Mechanical Radiation Detectors

In the preceding section it has been shown that the sensitivity of maser amplifiers is reduced at higher frequencies because of the noise temperature associated with spontaneous emission. At $\omega/2\pi \approx 10^{13} \text{ sec}^{-1}$ a maser amplifier can be expected to detect approximately 10^6 photons. At the same time it is known that the sensitivity of γ -ray counters or photomultipliers is great enough so that single photons can be detected.

It has been shown in reference 5 that the high sensitivity of counters is due to the fact that there is no noise due to spontaneous emission. In the same work a microwave detector free from noise due to spontaneous emission was proposed. The principle of operation of this detector is as follows (Fig. 22). Suppose there are $2j + 1$ equidistant energy levels, as might be found, for example in the interaction of the

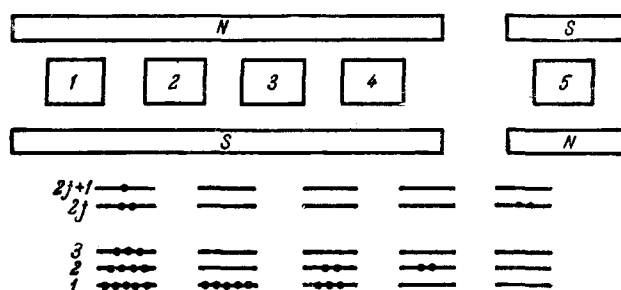


FIG. 22. Principle of operation of a quantum-mechanical detector. 1) source of molecules, 2) selection system, 3) resonator, 4) second selection system, 5) second resonator.

magnetic moment of a molecule with an applied magnetic field. By means of an inhomogeneous magnetic field, molecules in the lower energy level can be removed from the beam, which then passes through a cavity resonator tuned to the frequency corresponding to the transition between adjacent levels. The radiation to be detected enters the resonator and is absorbed by the molecules, so that a fraction of the molecules, proportional to the number of detected photons, is raised to the first excited level. A second selection system, located beyond the resonator, is used to isolate molecules in the first excited level. The molecular beam, processed in this way, then enters a region of space in which there is a magnetic field in the opposite direction to the original field. Under these conditions the energy levels are inverted so that only the second level (counted from the highest level) is populated. After a period of time this system radiates $(2j - 1)$ photons for each of the detected photons. Thus, this system "multiplies" the original photons by a factor of $(2j - 1)$. A system of this kind is sensitive to photons characterized by frequencies which lie within the width of the spectral line. Since the frequencies of the radiated photons are not related uniquely to the frequency of the absorbed photons, this system can be used as a detector, but not as an ampli-

fier. More precisely, whereas this system is capable of detecting photons over a relatively wide frequency range, because of the molecule-resonator interaction time the bandwidth of the system is determined by the lifetime for spontaneous emission in the excited state $\tau \approx 10^6$ sec for $\omega/2\pi \approx 3 \times 10^{10}$ sec⁻¹. Because the quantum-mechanical system is initially in the lowest energy state, this detector is free of spontaneous emission noise.

One shortcoming of this system is its complexity. In addition, the long lifetimes characteristic of the centimeter region make it unfeasible in practice.

Another system for detection of infrared and optical radiation in which there is no noise due to spontaneous emission is described in reference 6. Suppose that the quantum-mechanical system has the energy levels shown in Fig. 23. The frequency ω_{12} corresponds to the detected infrared radiation. There is pumping radiation at frequency ω_{32} which saturates levels E_2 and E_3 . At low temperatures only E_1 is populated. Photons which are absorbed at frequency ω_{12} cause transitions to E_2 ; then, because of the pumping radiation at frequency ω_{32} , transitions are caused from E_2 to E_3 . The subsequent transitions from E_3 to E_4 occur spontaneously and are detected with a photomultiplier because ω_{34} is an optical frequency. Between the system and the photomultiplier there is a filter which absorbs the optical pumping radiation at frequency ω_{32} .

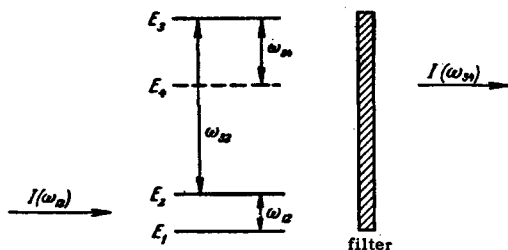


FIG. 23. Level diagram for an infrared detector.

Because the photomultiplier can detect single photons in the optical region the sensitivity of the system is very high.

At low temperatures the system is in the ground state E_1 so that there is no spontaneous emission noise. As in the preceding case, this system is not an amplifier. In reference 6 the system is called a quantum counter.

Analysis of the systems described above shows that the noise can be reduced by reducing the temperature. However, because of the statistical nature of spontaneous emission these systems have finite noise temperatures.

CONCLUSION

Within the short time they have been under development, quantum-mechanical systems have become extremely valuable for the generation and amplification

of electromagnetic waves in the centimeter and decimeter regions. The availability of maser oscillators and amplifiers in these regions has cleared the way for two important advances in radio engineering:

1) an appreciable increase in the frequency stability of oscillators and 2) a substantial increase in the sensitivity of detectors. The use of ammonia masers has already made it possible to achieve high frequency stability.⁶⁹ Maser amplifiers in the centimeter and decimeter wavelength ranges have noise temperatures of the order of 10°K. Maser oscillators and amplifiers have also been used to solve a number of scientific and technical problems in radar, navigation, long-distance radio communication, radioastronomy, the design of frequency standards (time), etc.

More than a thousand papers have already been published in this field; among these papers there are review articles^{1-3,10} and monographs^{70,71} which are concerned with the theory, design, and application of quantum-mechanical devices. This work comprises a new field of physics, which is now known as quantum electronics.

At the present time, research is under way in laboratories in many countries to explore the use of quantum-mechanical systems for the generation, amplification and detection of electromagnetic radiation in the submillimeter, infrared and optical regions.*

Many methods have been proposed for obtaining negative temperatures in different quantum mechanical systems and a number of theoretical problems have been treated. Most of this work has been described in the present review. However, it should be emphasized that amplification or oscillation in the infrared or optical wavelength regions has not yet been reported. It is apparent, however, for example from the present review, that there are no fundamental difficulties; it is reasonable to expect that infrared and optical oscillators and amplifiers will be built in the near future.

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Translator's Comments. A number of important developments in the optical maser field have been reported in the American literature since the appearance of this review by Basov, Krokhin and Popov. For the most part these developments represent the experimental realization of systems which have been proposed in the literature.

The experimental development which is perhaps of the greatest interest is the successful operation of a cw optical maser, which has been reported by Javan, Bennett and Herriott.¹ These authors have obtained population inversion and continuous maser oscillation in a gas discharge containing an He-Ne mixture. Evidence for the existence of a negative temperature in an Hg-Zn mixture has also been reported,² although experimental details are not available at the present writing. A theoretical paper which discusses negative temperatures in discharges in gas mixtures has also appeared recently.³

The work described in solid-state maser research has been concerned mainly with the pulsed ruby maser.⁴ Various features and modes of operation of this device have been described^{5,6,7} and a

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Translated by H. Lashinsky

theoretical analysis of some of these features has been given.⁸ Pulsed optical masers in which other materials are used have also been reported; the pumping power required with these materials is much lower than that needed for the ruby maser.^{9,10}

A further analysis of the resonant modes in an optical maser in which a Fabry-Perot system is used has been given by Fox and Li.¹¹

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